



## **Speciation analysis of iodine isotopes ( $^{127}\text{I}$ and $^{129}\text{I}$ ) in aerosol using sequential extraction and mass spectrometry techniques**

**Zhang, Luyuan; Hou, Xiaolin; Xu, Sheng; Fan, Yukun; Zhou, Weijian**

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## MARC X Final Book of Abstracts

The organizers of the Tenth International Conference on Methods and Applications of Radioanalytical Chemistry (MARC X) conference are pleased to provide the following summary of abstracts for reference by attendees of the MARC X conference. These materials are a supplement to the actual Program which provides complete details of the conference and sequence of presentations. However it will not include a copy of all the abstracts as it would be a very large publication. MARC X is expected to be the largest MARC conference with over 470 abstracts submitted.

The following 300 + pages provide the abstracts submitted for presentation at the MARC X conference as of March 15, 2015. The list will only be provided electronically and only a small number of hard copy abstract summary books will be available for reference at the conference at the desk. Individual copies will not be provided.

If you have any questions please contact the program chair, Sam Glover ([sam.glover@uc.edu](mailto:sam.glover@uc.edu)). Please visit the MARC website for additional details at [www.marcconference.org](http://www.marcconference.org).

Log: 105. **PHYSICS AND CHEMISTRY OF HEAVY ION ACTIVATION – TWO DECADES JOURNEY.** Susanta Lahiri, Saha Institute of Nuclear Physics, India.

It was more than fifteen years before we intended to produce some no-carrier-added (NCA) rare earth radionuclides by proton/alpha particle activation followed by the subsequent separation from the target matrix. Due to the similar chemical properties it became a colossal task to separate NCA radionuclides from the adjacent target radionuclides. To get rid of this monotonous similarity, heavy ion activation was planned which yielded greater difference in atomic number between target and product radionuclide, a more favorable situation for separation of NCA radionuclides from the target matrix. The heavy ion activation opened a new domain of neutron deficient radionuclides, with shorter and convenient half-lives for biological or clinical applications. Later we have extended our study to the entire Periodic Table and proposed numbers of heavy ion induced routes for production of clinically important radionuclides. The important examples include Cu-61; Mo-93m; Tc-93-96; Ru-97; Eu-145-147; Gd-147,149; Tb-149-153; Tm-163,165; Re-181,183; Au-192,193; At-208-210, and many more. We also developed simple radiochemical methods for separation of these NCA radionuclides from the target matrix. The journey includes many first, including excitation function studies using exotic beams like Be-9 for production of Tc and At radionuclides, etc. We introduced the concept of tracer packet where a group of NCA radioactive isotopes of some closely associated elements can be produced together with an objective to study some particular physical, chemical or biological system. We also kept an eye on the responsibility of radiochemists for application of greener techniques for separation of these radionuclides and combined accelerator-based chemistry with green chemistry mandates.

Log: 106. **FUKUSHIMA- A VIEW FROM THE OCEAN** . Buesseler, K.O. Woods Hole Oceanographic Institution.

The triple disaster of the March 11, 2011 earthquake, tsunami, and subsequent radiation releases at Fukushima Dai-ichi were unprecedented events for the ocean and society. This presentation will provide an overview of studies of Fukushima radionuclides in the ocean. Total releases from Fukushima are still not well constrained, with estimates for cesium-137 from atmospheric fallout and direct ocean discharge spanning 4 to 90 peta Bequerels, but most likely in the 15-30 PBq range. This source is smaller than any  $^{137}\text{Cs}$  remaining in the N. Pacific from global and close-in fallout from the 1960's, and of similar magnitude to the ocean releases of  $^{137}\text{Cs}$  from Sellafield, though greater than fallout that reached the ocean from Chernobyl. The fate of cesium is largely determined by its soluble nature in seawater, though uptake in sediments does occur via cesium's association with both detrital particles and biological uptake and sedimentation. A mass balance of cesium supply from the rivers and ongoing leakage from the nuclear power plants suggests that sediments may remain contaminated for decades to come near Japan. Although levels of cesium in the ocean and being released from Fukushima nuclear power plants four years later are many thousand times lower than in 2011, other isotopes such as strontium-90 are becoming of greater concern as they are elevated relative to cesium in the groundwater and storage tanks at the reactor site. Across the Pacific, ocean currents carrying Fukushima cesium are predicted to be detectable along the west coast of North America by 2015. Although models suggest cesium will be at levels well below those considered of human health concern, the public is worried about the lack of ocean monitoring. An update on Our Radioactive Ocean (<http://OurRadioactiveOcean.org> ) will also be given, a citizen scientist launched to monitor the arrival of Fukushima cesium along the North American west coast over the coming 2-3 years.

Log: 107. **GENERATION OF NEPTUNIUM-236 AND PLUTONIUM-236 FOR USE AS CHEMICAL YIELD TRACERS IN THE MEASUREMENT OF NEPTUNIUM AND PLUTONIUM.** Jerome, S.M.(1); Ivanov, P.(1); Larijani, C.(1); Parker, D.J.(2); Regan, P.(1,3). (1) National Physical Laboratory, UK. (2) University of Birmingham, UK. (3) University of Surrey, UK.

The accurate and traceable measurement of neptunium and plutonium radionuclides are carried out in support number of areas of interest that include: (i) the civilian nuclear fuel cycle, (ii) military use of nuclear technology, (iii) environmental impact, and (iv) nuclear forensics. The traceable and accurate measurement of neptunium and plutonium are complicated by the need for suitable yield tracers for these particular elements. In order to reliably prepare these radionuclides in useable quantities with suitable purity from charged particle irradiation of uranium targets, a knowledge of the formation cross sections of Np-236 and Pu-236 is required for the candidate nuclear reactions, along with the formation cross sections of potential contaminant radionuclides. There is data available for the candidate nuclear reactions, although there are contradictions in the data available. Furthermore, there is little information relating to the formation of impurity radionuclides, particularly neptunium-237, and this is of vital importance in selecting the most suitable production route. In this paper, we measure the formation cross sections for neptunium-236g, neptunium-236m (and hence plutonium-236), neptunium-237, neptunium-238 (and hence plutonium-238) and neptunium-239 (and hence plutonium-239) in the irradiation of uranium-236 and uranium-238 by protons in the energy range 20-25MeV. The cross section data will be derived from the irradiation parameters (time and beam current) and the activity measurements of neptunium-236g, neptunium-237, plutonium-236, plutonium-238 and plutonium-239. Details will be given of the irradiation techniques employed, the separation chemistry carried out to isolate neptunium and plutonium as well as the measurement strategies used to quantify the radionuclides of interest.

Log 108: **RADIOCHRONOMETRY: A HISTORICAL PERSPECTIVE.** Williams, R.W. Lawrence Livermore National Laboratory, USA.

Soon after the discovery of radioactivity and the recognition of elemental transformations, the pioneers of the science postulated that measurements of parent and daughter elements could be used to measure time. A thread will be followed from these early postulates, through the measurement of half-lives, the development of analytical instrumentation, the development of the geologic time scale, to modern radiochronometry and the measurement of the age of nuclear materials.

Log 109: **DEVELOPMENT OF HIGH RESOLUTION RADIATION AND RADIOISOTOPE IMAGING METHODS IN PLANT AND APPLICATIONS TO THE STUDY OF RADIO-CESIUM FOLLOWING THE FUKUSHIMA ACCIDENT.** Nakanishi, T.M. University of Tokyo, Graduate School of Agricultural and Life Sciences.

This talk demonstrates that radiation and radioisotopes (RI) are indispensable tools to study distribution and uptake manner of water and nutritional elements in a living plant, based on our research. In the case of water, neutron beam provides water specific images and application of  $^{15}\text{O}$ -labeled water revealed water circulation in a plant. That is, tremendous amount of water was flowing out from xylem tissue, which was regarded as a mere pipe to transport water. Since there is a limit in resolution of positron images, we developed real-time imaging system using conventional RIs, such as  $^{14}\text{C}$ ,  $^{32}\text{P}$ ,  $^{35}\text{S}$  etc. Two types of real-time RI imaging systems were developed, macroscopic and microscopic systems. In the case of the macroscopic system, the element absorption manner from culture solution or from soil to roots, kept in dark, and then to the up-ground part, kept in light, was visualized. The real-time gas fixation manner was also able to image applying  $^{14}\text{C}$ -labeled carbon dioxide. In the microscopic imaging, a fluorescent microscope was modified to get both radiation and fluorescent images. Applying these imaging systems,  $^{137}\text{Cs}$  uptake manner in a rice plant was studied to analyze the effect of Fukushima nuclear accident. The agricultural implication of radioactive Cs in Fukushima after the nuclear accident is also introduced.



Log: 110. **The use of  $\gamma$ -irradiation to graft natural films.** Lacroix, M.(1), Khan, R.(2), Senna, M.(3), Sharmin, N.(1), Salmieri, S.(1), Safrany, A(4) (1) INRS-Institut Armand-Frappier, Canadian Irradiation Centre, Research Laboratories in Sciences Applied to Food, (2) Bangladesh Atomic Energy Commission, (3) Radiation Chemistry Department, National Center, Egypt, (4) International Atomic Energy Agency, Vienna.

Different methods of polymer grafting using  $\gamma$ - irradiation were done for the development of newly functionalized biodegradable films, and some physico-chemical properties are described. Zein and poly(vinyl alcohol) (PVA) were  $\gamma$ -irradiated in presence of acrylic acid (AAc) monomer for compatibilization. Resulted grafted films had their puncture strength (PS = 37-40 N.mm<sup>-1</sup>) and puncture deformation (PD = 6.5-9.8 mm) improved for 30 and 50% PVA in blend, with 5% AAc under 20 kGy. Methylcellulose (MC)-based films were irradiated in presence of 2-hydroxyethyl methacrylate (HEMA) or silane, in order to determine the effect of monomer grafting on the mechanical properties of films. Results showed that grafted films using 35% monomer performed higher mechanical properties with PS values of 282-296 N.mm<sup>-1</sup> and PD of 5.0-5.5 mm under 10 kGy. Compatibilized polycaprolactone (PCL)/chitosan films were developed via grafting silane in chitosan films. Resulting trilayer grafted composite film presented superior tensile strength (TS = 22 MPa) via possible improvement of interfacial adhesion PCL/chitosan) when using 25% silane under 10 kGy. MC-based films containing crystalline nanocellulose (CNC) as a filling agent were prepared and irradiated in presence of trimethylolpropane trimethacrylate (TMPTMA) as a grafted plasticizer. Grafted films presented superior mechanical properties with a TS of 47.9 MPa and a tensile modulus (TM) of 1792 MPa, possibly due to high yield formation of radicals to promote TMPTMA grafting during irradiation. The addition of CNC led to an additional improvement of the barrier properties, with a significant 25% reduction of water vapor permeability (WVP) of grafted films.

Log: 111. **Applications of Irradiation Processing in Combination with Ozonation, Antimicrobial Coating and Modified Atmosphere Packaging to Assure the Safety of Fresh Vegetables.** Lacroix, M.1, Severino, R.1,2, Vu, K.D.1, Salmieri, S.1, Donsi, F.2, Kermasha, S.3, Ferrari, G.2,4: (1) Canadian Irradiation Centre, INRS-Institut Armand-Frappier; (2) University of Salerno; (3) McGill University.

The antimicrobial effect of modified atmosphere (MAP), ozone and  $\gamma$ -irradiation in combination with a bioactive coating was evaluated on broccoli florets and beans inoculated with *L. Monocytogenes*, *Escherichia coli* O157: H7 and or *Salmonella typhimurium*. Inoculated samples with pathogens, were treated with coating formulations based on palmitoylated chitosan (MC) containing nanoemulsions of carvacrol (CA) or mandarin essential oil (MA). The MA nanoemulsion was tested in combination with ozonated water and  $\gamma$ -ray, and the results showed a synergistic effect, increasing the *L. monocytogenes* radiosensitization by 1.25-fold, with the results of ensuring microbial safety over 13 days storage: the microbial load reduction was still of 2.5 log CFU/g of *L. monocytogenes* after 13 days. The

antimicrobial activity of the coatings containing nanoemulsion of carvacrol and  $\gamma$ -irradiation under MAP was then evaluated on green bean samples. The radiosensitivity of *E. coli* and *S. typhimurium* to  $\gamma$ -irradiation showed that, without MAP, MC-based coating containing carvacrol nanoemulsion significantly increased the radiosensitization of *E. coli* and *S. typhimurium* by 1.32-fold and 1.30-fold, respectively. The use of bioactive coating under MAP caused a synergistic effect with an increase in radiosensitivity by 1.80-fold and 1.89-fold for *E. coli* and *S. typhimurium*, respectively, and reduced the microbial population to undetectable levels during the whole storage for *E. coli* and from day 7 to the end of storage for *S. typhimurium*. The obtained results are of great interest to food companies aiming to ensure the safety of ready-to-eat food products with a prolonged shelf life.

Log: 112. **HANFORD RADIONUCLIDE SOURCE TERMS SERVE TREATY RESEARCH.** Wogman, N.A. Pacific Northwest National Laboratory (Retired - Emeritus Status).

Hanford U.S.A. radionuclide releases led to fate and transport research. These studies have been applied to research involving Treaties such as the Non-Proliferation Treaty (NPT) and the Comprehensive Nuclear Test Ban Treaty (CTBT). The outcomes are to develop sensor and environmental sampling technologies such that as much signature material as possible is removed from the largest volume/mass of environmental material followed by analysis on the lowest background/highest efficiency systems -- i.e., the endeavor is to collect as much source term as possible in the smallest sample with analysis on the highest efficiency and lowest background sensor system such that the lowest limit of detection is achieved. The International Atomic Energy Agency (IAEA) addresses the NPT while the CTBT is pursued by the CTBT Organization. Both require environmental sampling and radionuclide analysis covering the world's environs. The efficacy of long- and short-range monitoring for both the IAEA and the CTBT depends on the availability of mobile signature radionuclides and on the ability to distinguish them from background signals and attribute them to a source. Signatures released to the environment must be distinguished from natural backgrounds, old fallout materials, fuel reprocessing, and medical radioisotope production to name but a few. This paper serves as a scientific basis to start discussions of the available environmental sampling and sensor techniques that should be used within the context of monitoring for the detection of undeclared nuclear activities for the IAEA as well as for the detection of a nuclear test for the CTBT. This work contains information on the techniques, equipment, costs, and some operational procedures associated with wide-area environmental sampling as well as a discussion of issues, recommendations, and questions needing further study. The sampling and analysis of aquatic materials, atmospheric gases, atmospheric particulates, vegetation, sediments, soils, and fauna are also discussed.

**Log: 113. Indoor radon (Rn-222) concentration level study in childcare centers, kindergartens and elementary schools, using Nuclear Track methodology (NTM).** G. Espinosa and J.I. Golzarri.

As it is known, staying for long periods of time in a place with high concentration of radon is a health hazard. This health risk can be increased into the adulthood, if on infant ages the young people growing inside of a places with important indoor radon levels. Causal associations between exposure level and lung cancer have been demonstrated in epidemiological studies around the world, suggested that for younger age children groups, the risk coefficient of lung cancer for inhaled radon and their daughters is about a factor of 4 on children from recent born to 10 years old, and 2 for ages between 10 to 20 years old. Indoor radon (Rn-222) concentration levels were measured in 10% of the childcare centers, 8% of the kindergartens, and 8% of the elementary schools, in Ixtapalapa, a borough of Mexico City located in the eastern part of the city. The pilot study was conducted over a one-year period, divided into four three-month periods coinciding with the seasons. Nuclear Track Detection Methodology was selected for the measurements. A passive close-end cup device with CR-39 Lantrack® polycarbonate as the detection material was used, with one step chemical etching process, following a well established detection and analysis protocol originally developed by the "Dosimetry Applications Project" (DAP), of the Physics Institute of the National Autonomus University of Mexico. The indoor radon concentration levels ranged from 65 to 173 Bq/m<sup>3</sup> in the childcare centers, from 60 to 140 Bq/m<sup>3</sup> in kindergartens, and from 50 to 115 Bq/m<sup>3</sup> in elementary schools. It is important to point out that this study represents the first time in Mexico City, that indoor radon measurements have been carried out in childcare centers, kindergartens and elementary schools. The importance of the study is clear given the time that children (carers and teachers) spend in childcare centers, kindergartens and schools. This work was partially support by UNAM-DGAPA-PAPIIT project IN103013.

Log: 114. **Natural radioactivity determination of some traditional Mexican medicinal herbology, by gamma spectrometry with HPGe detector.** Espinosa, G., Golzarri, J.I., Chavarría, A.

In this work, the result of the gamma spectrometry analysis of 30 of the Mexican medicinal herbs is presented. The analysis was carried out using low level background shielding, a hyper-pure germanium (HPGe) detector, and a 16k Multichannel Analyzer (MCA) system. The Mēxihcah (Mexica/Aztecs) used herbs to treat illnesses typical of Mexico, beginning around 800 AC. Later on, after the Spanish conquest of Mexico in around 1520 AC, the information about these medicinal herbs contained in pre-Columbian Aztec codices, was transferred to other codices including the Codex Matritense, the Florentine Codex, and the Codex De La Cruz-Badiano. The last of the three known as: Amate Cehuatl Xihuitl Pitli in Nahuatl, and Libellus de Medicinalibus Indorum Herbis in Latin; and it is a pictorial compilation of the medicinal plants used in New Spain in the 16th century, and is considered to be one of the master pieces of the world literature. The natural radioactivity from the U-238, U-235, Th-232, and their daughter, and from K-40, were determinate by gamma spectrometry, with HPGe and MCA detection system as mentioned above. The Ortec-Maestro® for Windows® software was chose for the analysis, and calculations were carried out using MS-Excel®. The spectrometry system was calibrated for energies from 0.6 to 1.5 MeV. The Mexican herbology plants selected for this study, were certified by "Universidad Nacional Autonoma de Mexico (UNAM)" Medicine Faculty, and were collected during the year 2013. The plants studied were found to present low radioactivity levels; these levels cannot be considered a health risk for consumers. The present study is the first radioactive content analysis of the traditional Mexican medicinal herbology. The study thus complements the body of knowledge concerning the concentration of natural radioactive elements in plants. [This work was partially support by UNAM-DGAPA-PAPIIT project IN103013.

Log: 115. **Fabrication and Testing of a High Pressure Carbon Fiber Marinelli Vessel for the Measurement of Radioactive Gases.** Mann, N.R.(1); Robinson, T.A.(1); Watrous, M.G.(1); Oertel, C.P.(1); McGrath, C.A.(2) (1) Idaho National Laboratory. (2) Idaho State University.

The INL produces radioactive gas calibration standards in support of the CTBTO. While the precursors of these standards are quantified prior to mixing and packaging, no quantification method currently exists for the final unaltered product. Researchers at the INL have developed a pressure vessel in a standard Marinelli configuration, which will allow for the quantification of the final gas product without any alteration. Testing of aluminum/stainless steel prototypes has shown promise. However, the detection is limited by the attenuation of the materials of construction and the maximum working pressure of 1000 psi. INL has partnered with Composite Technology Development Inc. (CTD) to fabricate a high pressure analysis vessel in the standard Marinelli configuration. The partnership will utilize the composite carbon fiber materials to provide an overall benefit due to an increased operating pressure with decreased attenuation from the materials of construction. Unlike traditional composite vessels that comprise of a composite outer layer and metallic or polymeric inner liners, CTD has developed proprietary resin systems that create a linerless shell. The shell serves as both a permeation barrier, as well as providing the structure necessary to carry all pressure, thermal and structural loads. Preliminary carbon fiber composite designs estimate a working pressure five times greater than the current aluminum/stainless steel prototype. In addition, by eliminating the polymer and metallic liners within the composite shell, gamma energies will need only to pass through the low-Z carbon fiber and resin system. The results obtained with a carbon fiber Marinelli vessel are presented.

Log: 116. **Argonne's non-linearity/mass-bias correction equation for ICP-QMS.** Steeb, J.L.; Graczyk, D.G.

In following up on inconsistencies that we saw in measurements using the quadrupole ICP-MS on validation of an isotope-dilution procedure for age-dating Sr-90 sources, Argonne developed a new two-parameter modeling formula for normalizing data to pertinent isotopic reference materials. This methodology led to improving the short-term and long-term precision as well as accuracy of the measurements. On applying the model to other tasks, we found that values for the parameters, which account for non-linearity in the system response and mass-dependent variations among recorded signals, do not conform to expectations based on conventional interpretation. Within a single analysis run for a specific element, the two parameters can be evaluated to very tight tolerances and the model fits the data to within a few parts per thousand. However, in a different run or with a different element, the values of the two parameters are not the same. In this talk we will present our new modeling formula, our comprehensive strategy for conducting measurements of isotope ratios or trace-element concentrations, and supporting data demonstrating how the nonlinearity correction equation has improved the precision and accuracy achievable with the widely available ICP-QMS system.

Log: 117. **Safeguards Applications for Uranium Impurity Analysis Data.**  
Peńkin, M.V.; Boulyga, S.F. IAEA Department of Safeguards.

The International Atomic Energy Agency (IAEA) collects and analyses samples of materials involved in the conversion of uranium into nuclear-grade products to support the verification of States' declarations and to look for indications of possible undeclared materials and activities. Dedicated sampling protocols and equipment have been developed and implemented, aimed at ensuring sample representativeness and at preserving its integrity and authenticity. Concentrations of over fifty elements of interest are determined using Inductively-Coupled Plasma Mass Spectrometry, with some complementary techniques applied. To provide confidence in the derived conclusions, samples are analysed by multiple laboratories. Consistency of the results is addressed through the unified data reporting requirements, the use of common reference materials, and via inter-laboratory comparisons. Impurities found in processed uranium are useful for discerning a variety of characteristics: some of them are proven to be sustainable source-related signatures, others bear information about process reagents and construction materials; some ubiquitous elements are important parameters specified by industrial requirements. The impurity data, along with other essential parameters, are interpreted to judge sample conformity to the relevant specifications, to evaluate the facility design information, to assess material provenance and intended use, and to detect its possible substitution. Processing, organizing and evaluating multidimensional impurity analysis results require a well-structured database, established and reliable reference datasets, and complex mathematical tools. The IAEA has been using impurity information to aid in safeguards verification for several years, and will continue to explore and expand its use going forward.

Log: 118. **A RENAISSANCE OF PLUTONIUM METAL PRODUCTION AT THE GRAM SCALE.** R. A. Henderson, N. K. Harward, K. E. Roberts, J. A. McNeese, and K. J. M. Blobaum, Lawrence Livermore National Laboratory, 7000 East Avenue, L-236, Livermore, CA, 94550.

Normal production of plutonium metal is carried out on the kg scale, and has been done so since the early 1940's, once satisfactory methods were devised during the Manhattan project. Recent interest in low-activity, high-purity  $^{242}\text{Pu}$  components for physics experiments has led to the need to re-create metal production efforts on the gram scale due to the amount of target material available for the proposed experiments. We report on efforts to perform direct-oxide reduction at the gram scale, as well as efforts to re-create the conditions of the original metal production experiments performed some 70 years ago in Los Alamos. The final route to successful Pu metal production is described. A relatively recent work by Usami(1) showed the possibility of producing plutonium metal by direct-oxide reduction on the gram scale, a process that wasn't pursued by Baker(2) in the early 1940's at the Los Alamos National Laboratory. The Baker methods include thermite-type bomb-reductions using plutonium flourides as starting materials. These processes had not been attempted at the Livermore National Laboratory, so a series of experiments was undertaken to learn the techniques required for successful production of plutonium metal. The initial experiments were performed using cerium as a surrogate, and after vetting the process to be used, the reductions were again performed with plutonium.

Log: 119. **IAEA INTERLABORATORY PERFORMANCE TEST EXERCISE ON LOW-LEVEL PLUTONIUM ISOTOPE MEASUREMENTS IN BIOASSAY SAMPLES USING ACCELERATOR MASS SPECTROMETRY (AMS).** Hamilton, T.F.(1); Child D.(2); Llerandi, C.S.(3); Salbu, B.(4); Aragón, A.(3); Antón, M.P.(3); Brown, T.A.(1); Hotchkis, M.(2); Kehl, S.(1); Lind, O. C.(4); Martinelli, R.(1); Tumey, S.(1); Fesenko, S.(5). (1) LLNL, USA. (2) ANSTO, Australia. (3) CIEMAT, Spain. (4) Norwegian University Life Sciences, Norway. (5) IAEA, Austria.

A world-wide performance test exercise on low-level measurements of plutonium isotopes in bioassay samples was formulated and conducted under the auspices of the International Atomic Energy Agency (IAEA) Coordinated Research Project (CRP) (K4 1013). The test measurements focused on the use of accelerator mass spectrometry and involved participating laboratories from Australia, Norway, Spain and the United States. The test samples were prepared and distributed using standardized protocols developed for participants in the Intercomparison Studies Program (ISP) at the Oak Ridge National Laboratory (ORNL). Each laboratory received a total of 17 performance test samples including 12 samples containing natural urine and 5 bottle blanks with deionized water. The natural urine samples were spiked in triplicate and nominally contained between 0 and 100  $\mu\text{Bq}$  of Pu-239 and a Pu-240/Pu-239 atom ratio of 0.02 to 0.1. Each laboratory was asked to use their own standardized sample preparation and measurement technique, and provide electronic reporting of data to ORNL. This paper will discuss the results of this performance test exercise.

Log: 120. **USE OF BIOCHAR AS A SOIL ADDITIVE FOR REDUCING THE SOIL-TO-PLANT UPTAKE OF CESIUM ISOTOPES IN RADIOACTIVELY CONTAMINATED ENVIRONMENTS****SIN-SITU GEOSPATIAL MEASUREMENTS OF CESIUM-137 CONTAMINATION ON RONGELAP AND UTRŌK ISLANDS IN THE NORTHERN MARSHALL ISLANDS USING A MOBILE CARBORNE GAMMA-SPECTROMETRY SYSTEM.** Hamilton, T.F.(1); Martinelli, R.(1); Peters, S.(1); Kehl, S.(1); Tamblin, M.(1); Hayes, M.(2); Hawk, D.(3); Smith, I.(4); (1) LLNL, USA. (2) University of Limerick, Ireland. (3) Space and Earth Carbon Research Team, Oneida Indian Reservation, USA. (4) Northern Arizona University, USA.

Pyrogenic carbon (Cpyro), commonly known as Biochar or Terra Mulata, is a porous charcoal that has been derived from partial carbonization of ligno-cellulosic materials including plant and waste biomass through pyrolysis. The use of Biochar in agriculture dates back to 7000 BC forming part of the Ancient Amazonian Dark Earth soils distribution (Amazon Black Earth). The humic components derived from Black Carbon have high aromaticity and charge density giving rise to high CEC values and sustainable fertility. Biochar is extremely stable in soil and is known to greatly improve the productivity of plants. There is growing interest in Biochar research as a carbon-negative pathway for semi-permanent storage of atmospheric carbon dioxide. In this study, we have explored the remediation properties of woodlands tree Biochar as a potential soil additive to selectively sequester labile soil cesium or otherwise help reduce the uptake of cesium isotopes into plants. In a batch tracer experiment, fine grain fractionated Biochar was found to have a relatively high affinity for adsorption of Cs ions ( $K_d > 100$ ) relative to that observed in untreated coral soils from the Marshall Islands. It should also be noted that the Biochar used in this experiment contained a significant source of potassium and magnesium. Both these findings support our hypothesis that the use of Biochar as a soil additive may provide a simple yet effective method for reducing the soil-to-plant transfer of cesium isotopes in contaminated environments.



Log: 121. **IN-SITU GEOSPATIAL MEASUREMENTS OF CESIUM-137 CONTAMINATION ON RONGELAP AND UTRŌK ISLANDS IN THE NORTHERN MARSHALL ISLANDS USING A MOBILE CARBORNE GAMMA-SPECTROMETRY SYSTEM.** Hamilton, T.F.(1); Kehl, S.(1); Tamblin, M.(1); Dennison, D.(1,#). (1) LLNL, USA. (#) Jacobs Engineering Group, LLNL, USA.

Nuclear accidents and events such as the 2011 Fukushima Daiichi nuclear power station incident in Japan emphasize the need for rapid reporting of geospatial information on the deposition of radioactive contaminants in the environment. The Marshall Islands Program in cooperation with Radiation Solutions Inc. (RSI) has successfully deployed an advanced mobile gamma-ray spectrometry system for use in the Marshall Islands. The RSI measurement system consists of two 4 L sodium-iodide (NaI) detectors coupled to a (Photomultiplier Tube) PMT system mounted on the roof of a Rough Terrain Vehicle (RTV). The system offers fully integrated digital spectral analysis with data processing, and real-time geospatial surveying and mapping capabilities. Applications of this carborne gamma-ray spectrometry (CGS) system at nuclear impacted sites in the Marshall Islands include efforts to assess the non-homogeneous distribution of cesium-137 contamination in surface soil on Rongelap and Utrōk Islands. These studies also provide a clear demonstration of the effectiveness of the soil remediation programs and general cultural practices in helping reduce external gamma exposure rates in and around village and housing areas.

Log: 122. **DEVELOPMENT OF RAPID RADIOCHEMICAL SEPARATIONS OF AMERICIUM/CURIUM FROM NUCLEAR FORENSIC MATRICIES USING SOFT N-DONOR LIGANDS.** Higginson, M.A. (1); Thompson, P. (2) Marsden, O.J. (2) Livens, F.R. (1); Heath, S.L. (1);(1) University of Manchester, UK. (2) AWE, UK.

Analysis of americium/curium is essential for determining the origin and intended use of unknown radioactive materials in nuclear forensics, but requires separation of Am/Cm from complex matrices containing actinides, lanthanides, fission and activation products, and matrix elements. Such separations (in particular Am/lanthanide) are typically challenging, involving multiple columns and/or use of thiocyanate for Am extraction, which generates toxic wastestreams. Soft N-donor ligands showing high selectivity ( $SF_{Am/Eu} > 250$ ) for Am/Cm over lanthanide analogues (Eu) such as BTPHens and BTBPs have recently been identified. These ligands are promising for the development of a rapid selective americium/curium separation process from complex samples. We have identified and synthesised a range of triazine soft N-donor ligands and developed processes for the separation of Am/Cm from simulated nuclear forensic matrices. Using a combined method of AGI-X8 chromatography and liquid-liquid separation, selective extraction of americium was achieved from mixtures containing An, Ln, fission products, matrix elements and high-level Fe, Ca and Al. This procedure allows separation and quantification of all major actinides (U, Pu, Np and Am/Cm) within a 2 day timeframe with high decontamination factors ( $>1000$ ). Matrix elements are quantifiable by ICP-MS post-extraction with high recoveries ( $>90\%$ ). In addition, we have combined commonly-used extraction chromatography (EC) methods with our soft N-donor Am separation process for wider application within analytical radiochemistry. Immobilization of a BTPHen ligand onto a polymer support by covalent linkage has allowed development of an Am/Cm extraction resin as an alternative for liquid-liquid Am separation.

Log: 123. **DETERMINATION OF RADON DIFFUSION FROM SEDIMENTS VIA AN OPEN-LOOP APPROACH.** Burnett, W.C. (1); Chanyotha, S. (2); Kranrod, C. (2); Lane-Smith, D. (3). (1) Florida State University; (2) Chulalongkorn University; (3) DurrIDGE Co. Inc.

Radon flux via diffusion from sediments may be determined via a closed loop system but is complicated by the necessity of having to determine the total air volume in the system and accounting for any small air leaks that can arise over extended periods. We have now designed a simple laboratory system to evaluate radon diffusion from soils and sediments in the laboratory via an open-loop configuration. The setup includes a measured mass of wet sediment and water inside a gas-tight reaction flask connected to a drying system (active dry stick ADS-3) and a radon-in-air analyzer (DurrIDGE RAD7). Air flows through two activated charcoal columns before entering the reaction vessel to eliminate radon from the ambient air. The radon-free air then travels through a gas diffusion stone in the reaction flask, then through the ADS drier and finally to the radon analyzer where the radon activity is measured and recorded at regular intervals before the air is vented. After some time, the radon activity will reach a steady state depending upon the rate of radon diffusion from the sample and the airflow rate. Since the radon activity will increase with longer exposure times, it is advantageous to maintain a low airflow rate through the system. With this simple setup, the radon flux via diffusion is simply the product of the measured radon activity (Bq/m<sup>3</sup>) multiplied by the flow rate (m<sup>3</sup>/min).

Log: 124. **A MONTE CARLO APPROACH TO FOOD DENSITY CORRECTIONS IN GAMMA SPECTROSCOPY.** Metzger, R.L.(1); Van Riper, K. (2); Pouquette, P. (1). (1) Radiation Safety Engineering, Inc. (2) White Rock Science.

Many contaminating radionuclides that may be found in food products following a nuclear incident require a correction for the density of the food to obtain an accurate estimate of the specific activity of the contaminant using high resolution gamma spectroscopy. This correction is frequently obtained by using different multi-nuclide standards at different densities for the geometry of choice. Efficiency calibrations are obtained for each standard, and the sample results are corrected by interpolation between the appropriate efficiency curves. This approach is expensive in the cost of the multiple standards, time to obtain the efficiency curves, and finally in radioactive waste disposal. An alternate method has been developed by creating a detailed model of the HPGe crystal, germanium dead layer, cover, and sample container for the Monte Carlo transport code MCNP. The Monte Carlo code was then used to generate a series of efficiency curves for a wide range of sample densities. The method was validated by comparing the MCNP generated efficiency curves against those obtained from measurements of NIST traceable standards, and spiked food samples across a range of food densities and products. The Monte Carlo generated efficiency curves were found to be equivalent to the standard derived curves, and at far less cost.

Log: 125. **ISOLATION AND PURIFICATION OF THE XENON FRACTION OF  $^{252}\text{Cf}$  SPONTANEOUS FISSION PRODUCTS FOR THE PRODUCTION OF RADIO XENON CALIBRATION STANDARDS.** Houghton, T.P. (1), McGrath C.A. (2), Hague R.K. (1), Eisenmenger, J.G. (1). (1) Idaho National Laboratory. (2) Idaho State University.

The presence of radioactive xenon isotopes indicates that fission events have occurred, and is used to help enforce the Comprehensive Test Ban Treaty. Idaho National Laboratory (INL) produces  $^{135}\text{Xe}$ ,  $^{133\text{m}}\text{Xe}$ ,  $^{133}\text{Xe}$ , and  $^{131\text{m}}\text{Xe}$  standards used for the calibration and testing of collection equipment and analytical techniques used to monitor radio xenon emissions. At INL, xenon is produced and collected as one of several spontaneous fission products from a  $^{252}\text{Cf}$  source. Further chromatographic purification of the fission gases ensures the separations of the xenon fraction for selective collection. An explanation of the fission gas collection, separation and purification is presented. Additionally, the range of  $^{135}\text{Xe}$  to  $^{133}\text{Xe}$  ratio that can be isolated is explained. This is an operational update on the work introduced previously, now that it is in operation and has been recharged with a second  $^{252}\text{Cf}$  source.

Log: 126. **Self-attenuation Correction in Gamma Counting of Flame-Sealed Radioxenon Samples.** Robinson, T.A.(1). (1) Idaho National Laboratory.

There are many laboratories world-wide that monitor air for radionuclides that are indicative of nuclear detonations and other nuclear processes. Equipment that is operated for this purpose benefits from calibration with the same nuclides that are being monitored. Standards produced for calibration purposes are made regularly by INL for various interested parties using the commonly monitored short-lived xenon isotopes:  $\text{Xe-}^{131\text{m}}$ ,  $\text{Xe-}^{133}$ ,  $\text{Xe-}^{133\text{m}}$ , and  $\text{Xe-}^{135}$ . During standard production and quantification, a potential source of bias for the final activity concentration arises from attenuation of emitted gamma-rays by the sample gas itself. This bias effect has previously been ignored in standard quantification under the assumption that this effect was negligible. This work estimates the magnitude of the self-attenuation through modeling and quantifies the self-attenuation radioanalytically by studying the lowest energy gamma emitting radioxenon isotope used in gas standard production:  $\text{Xe-}^{133}$ . The studied radioactive xenon sample configuration consists of xenon gas that is flame sealed in borosilicate glass ampoules and counted by HPGe well detector. Results indicate that self-attenuation is quantifiable and, while lower in magnitude than the total uncertainty of produced standards, is a contribution of bias in our process that can now be removed with the information presented here. Application of the self-attenuation correction during calibration and sample quantification will also be presented.

Log: 127. **Separation of High-Purity Plutonium-244 for Safeguards Applications.** Penkin, M.V.(1); Humphrey, M.A.(1); Kryzhanovsky A.A.(2); Vyachin V.N.(2); Iyengar A.(3). (1) IAEA Department of Safeguards. (2) Russian Federal Nuclear Center - Institute of Experimental Physics. (3) US Department of Energy.

Since its inception, environmental sampling has proven to be one of the most valuable technical measures introduced to strengthen international nuclear safeguards. Analysis of minute amounts of nuclear material in such samples requires highly-enriched isotopic tracers. Pu-244 is commonly recognized as the most suitable spike for low-level mass-spectrometric measurements of plutonium, allowing for accurate determination of all isotopes from Pu-239 to Pu-242. However the available reference materials based on Pu 244 are very few and lack isotopic purity. The need for highly-enriched Pu-244 was identified by IAEA in the early 1990s. In 2005, the US DOE agreed to supply a small quantity of plutonium dioxide containing about 17.5 at.% Pu-244, for two-phase electromagnetic separation at VNIIEF (Russia). After the feed material containing 0.5 g Pu was delivered to VNIIEF in 2012, a first round of separation yielded approximately 10 mg plutonium with about 98.9 at.% Pu-244. This intermediate product was sampled by IAEA, verified for isotopic purity and plutonium amount through analysis by two independent laboratories in Russia and the United States, and later used to feed the second round of separation which is believed to have produced 0.8 mg of plutonium with over 99.98 at.% Pu-244 - subject to the ongoing verification measurements. The product will be certified as a reference material, to meet the need of the IAEA Network of Analytical Laboratories for decades to come. The authors acknowledge the efforts of David Donohue, Stanislav Vesnovskii, and Steven Goldberg who contributed to the eventual success of this long-standing project.

Log: 128. **DETERMINATION OF FE IN BLOOD: AN ALTERNATIVE FOR SPORT MEDICINE.** Kovacs, L.(1); Zamboni, C.B.(1); Metairon, S.(1); Macedo, D.V.(2); Rizzutto, M.A.(3). (1) Instituto de Pesquisas Energéticas e Nucleares, IPEN – CNEN/SP. (2) Universidade Estadual de Campinas, UNICAMP. (3) Universidade de São Paulo, IFUSP.

In this study, we intend to present the details of a new clinical methodology for Fe blood evaluation using a portable X-Ray Fluorescence Spectrometry (PXRFS). The main advantage for using this portable instrument is the fast procedure for iron analysis in blood (few minutes) combined with the viability to use small quantities of blood (10 times less, at least) compared with the conventional clinical tests. Iron is a cofactor in a variety of cellular processes essential for athletic performance. High aerobic activity and dietary habits may results in depletion of body iron stores, which could decreases the aerobic performance increasing the risks of fatigue and immune disorders. Athletes, particularly those involved in endurance sports, are commonly diagnosed with iron deficiency. In this study, the Fe status was determined in whole blood of amateur athlete's using the PXRFS. The blood samples were collected at Laboratório de Bioquímica do Exercício (LABEX/UNICAMP - Brazil). In the study 18 male amateur athletes participated. These results were compared with the control group (subjects of same age but not involved with physical activities) and showed significant differences. The applicability of the

portable spectrometry showed to be appropriate for this clinical purpose. Our data can be useful to a well-planned nutritional proposition that could contribute to performance of endurance athletes.

Log: 129. **TRACKING INORGANIC ELEMENTS IN GRMD BLOOD DOGS SUBMITTED TO HASCS INVESTIGATED BY NAA TECHNIQUE.** Metairon, S.(1); Zamboni, C.B.(1); Suzuki, M.F.(1); Bueno Jr, C.B.R.(2); Andrade, T.O.(2); Landini, V.(2); Cangussu, E.B.(2); Caetano, H.V.A.(2); Zatz, M.(2). (1) Instituto de Pesquisas Energéticas e Nucleares, IPEN – CNEN/SP. (2) Centro de Estudos do Genoma Humano, Instituto de Biociências, USP.

Nowadays, many promising therapeutic strategies have been developed in animal models with Duchenne Muscular Dystrophy (DMD). An animal model which has a phenotype similar to patients with DMD has been bred in Brazil: Golden Retriever Muscular Dystrophy dogs (GRMD). Recently, the Human Genome Research Center (Biosciences Institute in Brasil) has shown that human adipose derived from stromal cells (hASCs) and injected systemically into GRMD dog cephalic vein paw are able to reach, engraft, and express human dystrophin in the host GRMD dystrophic muscle up to 6 months after transplantation, which improves the functional performance of injected animals without any immunosuppressant. In this study, the elements Br, Ca, Cl, Mg, K, Na, Fe, Zn and S were investigated due to the relevance for evaluation of the transplantation process. The elements concentrations were determined using NAA technique. The blood collection was performed before, during and after beginning the hASCs treatment. To perform this investigation, blood samples from nineteen GRMD dogs (4 Control, 7 Carrier and 8 Affected), that were breeding at Biosciences Institute/USP (São Paulo, Brasil), were irradiated in the IEA - R1 nuclear reactor at IPEN - CNEN/SP (Brasil). These data are the first results for comparing the progress of hASCs treatment in GRMD animal model. The knowledge of inorganic element concentrations in blood of GRMD dogs may help to evaluate the progress of hASCs treatment in this animal model, before tests being performed in patients with DMD.

Log: 130. **Rapid determination of long-lived radionuclides using accelerator mass spectrometry measurement without chemical preparation.** Hou, Xiaolin (1, 2); Liu, Qi (2); Zhou, Weijian (2). (1)Technical University of Denmark, Center for Nuclear Technologies, Risø Campus, 4000, Roskilde, Denmark. (2)Xi'an AMS Center, SKLLQ, Institute of Earth Environment, CAS, Xi'an, China.

Due to sufficient high abundance sensitivity and the feature of free isobaric interference, accelerator mass spectrometry (AMS), as the most sensitive method for the determination of isotopes, especially low level radionuclides, has been widely applied in many fields, such as archeology, geology, environment, and biomedicine, and nuclear safety. In the conventional AMS method, the element of radionuclide has to be separated from sample matrix and prepared in a small target, which involves time consuming chemical separation, takes a few days for sample preparation, and therefore is the major bottleneck limiting its application, especially in rapid analysis of radionuclides in the emergency situation. In this work, we present a novel AMS analytical method for rapid determination of radionuclides. In this method, the dried and homogenized sample is directly mixed with conductive material and pressed into target holder, which is used for AMS measurement. In this case, the entire analytical time can be significantly shortened from days to less than 1 hour. Determination of  $^{129}\text{I}$  and  $^{14}\text{C}$  in different environmental and biological samples such as soil, sediment, animal tissue, and vegetation, using the developed method will be demonstrated. For  $^{129}\text{I}$  determination, the sample of about 3-10 mg spiked with 0.5-1 mg  $^{127}\text{I}$  carrier is mixed with niobium powder, pressed into copper target hold, and then directly measured using a 3MV AMS for 10-20 minutes to get  $^{129}\text{I}/^{127}\text{I}$  atomic ratio. The similar procedure is also used for the determination of  $^{14}\text{C}$ , but no stable carbon is spiked when biological sample is used. The analysis of some certified reference materials for  $^{14}\text{C}$  and  $^{129}\text{I}$  shows the developed method is accurate and suitable for analysis of environmental and biological samples; this will be very useful for emergency analysis, as well as analysis of large number of biological and environment trace research.

Log: 131. **ESTABLISHING A NATIONAL NUCLEAR FORENSICS LIBRARY: KNOWLEDGE OBTAINED FROM THE RADIOLOGICAL SEALED SOURCE LIBRARY.** JODI L. CANADAY (1); DAVID B. CHAMBERLAIN (1). (1) ARGONNE NATIONAL LABORATORY.

A National Nuclear Forensic Library (NNFL) contains forensics signatures of nuclear and radiological materials used, produced or stored within the country. In the event of lost, stolen, or interdicted materials, the NNFL is a beneficial tool for answering the question "Is this our material?" in a timely manner; however, the development process for an NNFL can be daunting. The NNFL development process can be broken down to determine; type and level of information necessary, data collection techniques and organization of data for prompt and accurate analysis. Argonne National Laboratory, in collaboration with Idaho National Laboratory, has been successful in establishing the radiological materials component of a NNFL. The library does not track individual sources or users of these sources, but focuses on design details and the companies that produce and distribute these sources. The Argonne/INL Radiological Library contains a wealth of information on radiological sealed sources; however, not all of its contents is required to build a functional NNFL. In this presentation, we will discuss lessons learned from developing a radiological sealed source library; including the required data and attributes field necessary to build a functional NNFL, and how to repurpose and supplement data collected from other regulatory sources.



Log: 132. **USING RADON AS ENVIRONMENTAL TRACER FOR SUBSURFACE NAPL CONTAMINATION AND IN-SITU REMEDIATION ASSESSMENT.** Schubert, M. (1) (1) UFZ - Helmholtz Centre for Environmental Research; Permoserstr. 15; 04318 Leipzig; Germany.

Soil and groundwater contamination with non-aqueous phase liquids (NAPLs) has to be considered a major environmental problem worldwide. Effective clean-up measures necessitate the application of environmental tracers that allow localizing and quantifying the contamination prior to the remediation activities. The use of such tracers is also beneficial during the clean-up measure for the monitoring of the remediation success. Radon ( $^{222}\text{Rn}$ ) has proven to be a suitable tracer/indicator for the evaluation of subsurface NAPL contamination in both soil and aquifer. If compared to other applicable artificial tracers its main advantages are (i) its ubiquitous occurrence in soil gas and groundwater allowing its large-scale and long term use as tracer, and (ii) its straightforward and precise on-site detectability. The suitability of radon for NAPL contamination assessment and remediation monitoring arises primarily from its distinct water/air/NAPL partitioning behaviour. If residual NAPL is present in the subsurface, radon that is naturally present in soil gas or groundwater partitions preferably into the organic phase leading to a significant radon deficit in soil gas or groundwater within the contaminated area. The magnitude of the radon deficit compared to its local natural background concentration allows localizing subsurface NAPL contamination as well as evaluating the success of an in-situ remediation measure. The presentation will discuss the applicability of radon as environmental tracer for subsurface NAPL contamination and in-situ remediation assessment by summarizing its chemical/physical properties related to the subject, and by discussing a few exemplary site studies including both contamination assessment and monitoring of remediation activities.

Log: 133. **UNDERSTANDING TRITIUM SPECIATION IN NUCLEAR SITE MATERIALS FROM INCREMENTAL THERMAL EVOLUTION PROFILES.** Daeji Kim(1), Ian W Croudace(2), Phillip E Warwick(3).

Understanding the association and retention of tritium in metals has significance in nuclear decommissioning programs and can lead to waste reduction and recycling of materials. To develop insights, a range of metals from two nuclear sites and one non-nuclear site were investigated which had different exposure histories. Tritium speciation in metals was inferred through incremental heating experiments over the range 20-900°C using a Raddec Pyrolyser instrument. Systematic differences in thermal desorption profiles were found for non-irradiated and irradiated metals. In non-irradiated metals (e.g. stainless steel and copper) it was found that significant tritium could be incorporated following prolonged exposure to tritiated water vapour (HTO) or gases in nuclear facilities. This externally derived tritium enters metals by diffusion with a rate controlled by the composition and the surface character of the metal (e.g. painted, unpainted). The tritium is normally trapped in hydrated oxides lying along grain. In irradiated metals an additional type of tritium can form internally through neutron capture reactions. The amount formed depends on the concentration and distribution of traces Li and B in the metal as well as the integrated neutron flux. Liberating this kind of tritium typically requires temperatures above 800°C. The pattern of tritium evolution derived from simple thermal desorption experiments allows reliable inferences to be drawn on the likely origin, location and phases that trap tritium. Any weakly bound tritium liberated at temperatures of ~100°C is indicative of mostly HTO interactions in the metal. Any strongly bound tritium liberated from 600-900 °C is indicative of neutrogenic tritium formed via neutron capture by trace Li and B. The time required for efficient thermal desorption of tritium ultimately depends on the metal composition, its tritium exposure history, integrated neutron flux, sample size, sample geometry, heating rate and final desorption temperature. The results of desorption studies can be used to guide waste reduction, remediation and possible recycling.

Log: 134. **RAPID DETERMINATION OF STRONTIUM-90 FROM YTTRIUM-90 IN SEAWATER.** M. TAYEB (1,2), X. DAI (1), E.C. CORCORAN (2), D.G. KELLY (2). (1) CANADIAN NUCLEAR LABORATORIES LIMITED. (2) ROYAL MILITARY COLLEGE OF CANADA.

A RAPID METHOD FOR INDIRECT DETERMINATION OF  $^{90}\text{Sr}$  FROM ITS PROGENY  $^{90}\text{Y}$  IN SEAWATER HAS BEEN DEVELOPED. THE METHOD IS SUITABLE FOR THE DETERMINATION OF  $^{90}\text{Sr}$  IN SEAWATER FOR ROUTINE AND NON-ROUTINE MONITORING PROVIDED THAT THE FOLLOWING CONDITIONS ARE SATISFIED: (I) THERE IS NO  $^{91}\text{Y}$  PRESENT IN THE SEAWATER AND (II) SECULAR EQUILIBRIUM BETWEEN  $^{90}\text{Sr}$  AND  $^{90}\text{Y}$  IS ESTABLISHED. THIS RAPID METHOD EMPLOYS A FAST AND EFFECTIVE TWO-STEP PRE-CONCENTRATION TECHNIQUE THAT UTILIZES CALCIUM CARBONATE,  $\text{CaCO}_3$ , AND HYDROUS TITANIUM OXIDE (DENOTED AS HTIO) FOR THE CO-PRECIPITATION OF BOTH Sr AND Y IN SEAWATER FOLLOWED BY SEPARATION OF Sr/Y USING EXTRACTION CHROMATOGRAPHY EMPLOYING DGA-N RESIN COLUMNS. THE EFFECTIVENESS OF THE CO-PRECIPITATION TECHNIQUE USING HTIO IS REFLECTED BY THE MANAGEABLE PRECIPITATE MASS THAT IS FREE OF THE EXCESSIVE DOMINANT SALTS OF SEAWATER. THIS NOT ONLY ALLOWS FOR EASE OF SAMPLE HANDLING DURING RADIOCHEMICAL SEPARATION PROCEDURE, BUT ALSO ENABLES SEQUENTIAL MEASUREMENT OF  $^{90}\text{Y}$  BY BOTH ČERENKOV COUNTING AND LIQUID SCINTILLATION ASSAY (LSA) TECHNIQUES AFTER  $^{90}\text{Y}$  IS ISOLATED FROM THE MATRIX. ADDITIONAL ROBUSTNESS OF THE METHOD INCLUDES HIGH Y CHEMICAL RECOVERY (AVERAGE  $88 \pm 7\%$ ) AND SHORT SAMPLE PREPARATION TIME ( $< 4$  HOUR) FOR A SET OF 10 SAMPLES. IN THIS STUDY, CHEMICAL RECOVERY WAS MONITORED USING BOTH STABLE Y AND SR AS WELL AS RADIOTRACERS  $^{88}\text{Y}$  AND  $^{85}\text{Sr}$ .  $^{85}\text{Sr}$  WAS TRACED USING ITS GAMMA-RAY EMISSION AND  $^{88}\text{Y}$  WAS TRACED USING BOTH ITS GAMMA-RAY EMISSION AND ELECTRON CAPTURE (EC) DECAY. SAMPLES WERE MEASURED FOR  $^{90}\text{Y}$  BY ČERENKOV COUNTING IMMEDIATELY AFTER SEPARATION AND SUBSEQUENTLY BY LSA. THE AGREEMENT BETWEEN THE TWO COUNTING TECHNIQUES AND BETWEEN CHEMICAL RECOVERIES USING BOTH STABLE ISOTOPES AND RADIOTRACERS WAS SATISFACTORY. THE MINIMUM DETECTABLE CONCENTRATION (MDC) OF  $^{90}\text{Y}$  MEASURED BY ČERENKOV COUNTING AND LSA TECHNIQUES WERE  $0.11 \text{ Bq L}^{-1}$  AND  $0.18 \text{ Bq L}^{-1}$ , RESPECTIVELY, FOR A 1L SEAWATER SAMPLE AND A COUNTING TIME OF 1 HOUR ON A LOW BACKGROUND HIDEX LIQUID SCINTILLATION COUNTER (LSC). METHOD PERFORMANCE RESULTS DEMONSTRATED VERY GOOD AGREEMENT BETWEEN EXPECTED AND MEASURED ACTIVITY CONCENTRATIONS. ALSO, EXCELLENT LINEARITY FOR  $^{90}\text{Y}$  BY ČERENKOV COUNTING AND BY LSA TECHNIQUES OVER AN ACTIVITY RANGE FROM  $0.58 \text{ Bq L}^{-1}$  TO  $\sim 94 \text{ Bq L}^{-1}$  WAS ESTABLISHED.

Log: 135. **A COMPARISON OF THE RADIATION CHEMISTRY OF SEVERAL DIGLYCOLAMIDES.** Mincher, B.J.(1); Zarzana, C.A.(1); Groenewold, G.S.(1); Mezyk, S.P.(2); Wilden, A.(3); Modolo, G.(3) (1) Idaho National Laboratory. (2) California State University, Long Beach. (3) Forschungszentrum Juelich.

The diglycolamides are receiving a great deal of attention as potential reagents for actinide and lanthanide solvent extraction in both the American and European fuel cycle programs. The best studied are tetraoctyldiglycolamide (TODGA) and its isomer tetraethylhexyldiglycolamide (TEHDGA). Recently, derivatives of TODGA have been prepared that are designed to minimize third phase formation during metal loading of the solvent. These methylated TODGAs are designated MeTODGA, and Me<sub>2</sub>TODGA. To be useful in actual application these compounds must be stable in a high radiation environment. Here we have gamma-irradiated all four compounds under identical conditions; either in dodecane solution, or in dodecane contacted with varying amounts of nitric acid. The G-values for radiolytic degradation were measured, and the main decomposition products identified by liquid chromatography-electrospray time-of-flight mass spectrometry. The insights into the radiation chemistry of the DGAs gained by these comparisons are presented.

Log: 136. **HIGH QUALITY ACTINIDE TARGETS.** Loveland, W; Oregon State University.

The continuing interest in studying the synthesis of the heaviest elements and the renaissance of interest in nuclear fission has led to increased demand for high quality actinide targets for the study of charged particle and neutron induced reactions. Among the challenges presented to target makers are the need for high specific activity targets (1-500 MBq), targets with multi-nuclide deposits on them (<sup>235</sup>U, <sup>238</sup>U, <sup>239</sup>Pu on the same target), target purity (no hydrogen or a well-known amount of hydrogen in the actinide deposit), the need to characterize fragment energy loss in the target prior to use and the need to have no non actinide impurities in the target. In the course of preparing targets for the fission TPC and our own fission and heavy element research programs, we have encountered and solved these problems. I will describe: (a) the use of beams of <sup>252</sup>Cf fission fragments to measure fragment energy loss and target non-uniformities (b) the problems in placement and masking of multi-nuclide deposits on a single target (c) the problem of SiO<sub>2</sub> and other impurities that degrade fission fragments in the target deposits (d) our procedures to make <sup>239</sup>Pu targets by vacuum volatilization (e) the aqueous synthesis of actinide tetrafluorides and (f) issues with target shipment and storage.

Log: 137. **Assessment of Radioactive elements in canned tuna marketed in Saudi Arabia After Fukushima Accident.** Alkhomashi, N.(1); Alkhorayef, M.A.(2, 3); Alyousif, A. A.S. (1); Alotibi, W.(1). (1)KACST. (2)King Saud University. (3)University of Surrey.

Studies of radioactivity in seafood are important for estimating the radiological hazards to and dose exposure of the public. The importance of such studies has increased after Japan's devastating 2011 earthquake and the subsequent nuclear

accident at the Fukushima Daiichi power plant. In the current study, 32 brands of canned tuna that were purchased in supermarkets in Riyadh, Saudi Arabia were analysed to quantify the activity levels of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{40}\text{K}$ . These samples were processed in Japan, the Philippines, Thailand, Europe and the Middle East in 2011 and 2012. The samples were analysed using high-resolution gamma-ray spectroscopy. The highest activity concentrations of all three radionuclides were obtained in the samples that were processed in Japan in 2011 a few months after the Fukushima Daiichi accident. However, the activity concentrations of radiocaesium in all the samples in this study were far below the limit for food and beverages of 1,000 Bq/kg, which is the standard limit in the UK and Canada.

Log: 138. **THE SOLVENT EXTRACTION OF AM(VI) USING CENTRIFUGAL CONTACTORS.** Mincher, B.J.(1); Tillotson, R.D.(2); Law, J.D.(2); Schmitt, N.C. (2). (2) Idaho National Laboratory.

One of the major challenges in nuclear fuel cycle research is the development of a separation that can be deployed on a process scale for americium from the lanthanides. Unlike most lanthanides, americium can be oxidized to higher oxidation states, and this presents an opportunity to exploit high valent americium for such a separation. Strong oxidizing agents are required. However, the preparation of hexavalent americium using sodium bismuthate has now become routine in our laboratory. The success of separations based on oxidized americium depends on not only the yield of the higher oxidation state, but also its persistence in solution. In batch contact work, short contact times allowed for the extraction of Am(VI) with  $\text{DAm} \sim 4$ , using 1 M diamylamylphosphonate as the ligand. Based on bench-scale extraction and stripping studies a flowsheet was designed and a hot test was performed in a radiological fume hood using three CINC, 5-cm centrifugal contactors. Stage 1 was the americium extraction stage. In stage 2, americium stripping was attempted after reduction with a low concentration of hydrogen peroxide. As the only lanthanide that oxidizes and extracts in this flowsheet, it was attempted to reduce and strip cerium in the third stage contactor. The results of this hot test are compared with batch extraction studies, where it was found that americium extraction was comparable to batch contacts, while stripping was less efficient than expected. Proposals for the next test are discussed.

Log: 139. **PU ISOTOPES IN SOILS FROM THE DOWNWIND AREA OF LOP NOR NUCLEAR TEST SITE: REGIONAL FALLOUT VS. GLOBAL FALLOUT.** Bu, W.T. (1,2); Guo, Q.J. (1); Zheng, J. (2); Uchida, S. (2). (1) School of Physics, Peking University. (2) Research Center of Radiation Protection, National Institute of Radiological Sciences. Pu isotopes in the downwind areas of nuclear test sites should be investigated for radioactive source identification and radiological assessment. For the first time, soil core samples from Jiuquan region in the downwind area of Lop Nor nuclear test site (CNTs) were analyzed for Pu isotopes. The  $^{239}+^{240}\text{Pu}$  activities and  $^{239}+^{240}\text{Pu}$  inventories were obtained. The high Pu inventories in most of the sampling places revealed that this region was contaminated by the regional fallout Pu from the CNTs. The deposition of Pu from the CNTs in the downwind area was heterogeneous as the Pu inventories showed large variations. The contributions of the CNTs to the total Pu in the soils were estimated. The averaged  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio for all the soil samples was around 0.16, slightly lower than that of global fallout. This atom ratio could be considered as a mixed fingerprint value representing Pu isotopes from the CNTs. Pu in the soils of Jiuquan region showed faster downward migration rate compared with other investigated places in China.

Log: 140. **Study on sorption of Cs from aqueous solution by microcapsules enclosing natural minerals.** Chuan-Pin Lee (1); Ming-Chee Wu (1); Tsuey-Lin Tsai (2); Shih-Chin Tsai (3); Yu-Lin Tu (1); Chun-Hua Pan (1). (1)Department of Earth Sciences, National Cheng Kung University, Tainan 70101, Taiwan.(2)Chemistry Divisions, Institute of Nuclear Energy Research, Taoyuan 32546, Taiwan.(3)Nuclear Science and Technology Development Center, National Tsing Hua University, Hsinchu 30013, Taiwan.

It has been proved that geological minerals presented a favourable environment for sorption of Cs, such as zeolite, illite, montmorillonite, and kaolinite. Zeolite, a natural mineral ( $\text{Na}_8(\text{AlO}_2)_8(\text{SiO}_2)_{40} \cdot 20\text{H}_2\text{O}$ ), exchanger having high selectivity toward Cs were encapsulated in the biopolymer matrices (sodium alginate, NaALG). The characterization of microcapsules (MCs) enclosing natural minerals was examined by SEM-EDS, and the sorption of Cs were investigated by batch methods. Spherical and elastic granules were obtained, and the particle size of microcapsules was about 700  $\mu\text{m}$ . A relatively high sorption ratio of Cs above 90% for MCs enclosing zeolite was obtained in the presence of 0.1-6 M  $\text{HNO}_3$  and 0.001-1 M NaCl solution, and the equilibrium was attained within 5 h. The sorption isotherm of Cs followed a Langmuir-type equation. The relatively larger  $K_d$  value (120 and 50  $\text{cm}^3/\text{g}$ ) for Cs showed a selective sorption property in the presence of various metal ions under 2.5 M  $\text{HNO}_3$  and 1 M  $\text{NaNO}_3$ .

Log: 141. **Indoor External and Internal Exposure due to Building Materials containing NORM in Korea.** Mee Jang(1);Kun Ho Chung(1);Young Yong Ji(1);Jong Myong Lim(1);Chang Jong Kim(1);Mun Ja Kang(1);Guen Sik Choi(1).(1)Korea Atomic Energy Research Institute.

We are continuously exposed to ionizing radiation from Naturally Occurring Radioactive Materials(NORM). Residues arising from the NORM industry or from other industrial processes are quantitatively the major part of NORM. Much of residues are reused as building materials all over world. But, building materials containing NORM with excessive natural background play a significant role in indoor external and internal exposure. The increased tendency in the building material industry to use NORM residue increase the possibility to increase exposure by them. Therefore, to protect the public from natural radioactive material, it has established the "Act on safety control of radioactive rays around living environment" in Korea. In this regulation, there is an annual effective dose limit of 1mSv for building materials, but the activity concentration limit is not established yet. This paper reviews the current state of the natural radionuclides activity in building materials such as phosphogypsum board, tile and bricks used in Korea. And, in order to get activity concentration limit, we estimated the potential external exposure from radium, thorium and potassium using MCNP code and measured activities. Also we calculated the indoor radon concentration from the radium activity and evaluated its effect to activity concentration limit.

Log: 142. **IN-BEAM NEUTRON ACTIVATION ANALYSIS AT MLZ (GARCHING).** Revay, Zs., Kudejova, P., Kleszcz, K Heinz Maier-Leibnitz Zentrum - Technische Universitaet Muenchen, FRM II.

The prompt gamma activation analysis (PGAA) facility of MLZ located at the FRM II, TUM, Garching operates the strongest cold neutron beam and has been successfully applied for the analyses of samples with masses less than a milligram, as well as for the investigation of low-cross-section nuclides and even radioactive targets. Recently, a low-background counting chamber has been added to the PGAA instrument thus enabling the counting of induced radioactivity. Since the beam flux is almost as high as that in smaller reactors ( $6 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$ ), it can efficiently be used for traditional neutron activation analysis not just in the case of short or medium-lived isotopes, but also for long-lived ones. As it is well-known PGAA and NAA complement each other, i.e. the first method provides the matrix composition, while the second one the trace elements, however their common potential has just partly been exploited. The combined analytical tool has been tested with standard and other reference materials, and successfully applied for a set of samples in material science etc.

Log: 143. **A FEASIBILITY STUDY ON THE PRODUCTION OF 88-Zr/88-Y GENERATORS BY PROTON BOMBARDMENT OF TANDEM TARGETRY.** Steyn, G.F.(1); Szelecsényi, F.(2); Kovács, Z.(2). (1) iThemba LABS, Faure, South Africa. (2) ATOMKI, Debrecen, Hungary.

The radionuclide 88-Y (half-life 106.6 d) has already found application in many practical fields. In nuclear medicine, for example, it can be used as a substitute for the therapeutically relevant but weak gamma emitter 90-Y, as a tracer for determining chemical yield and labeling efficiency. Besides the direct production via the (p,n) reaction on 88-Sr, it can also be obtained via the 88-Zr/88-Y generator. The 83.4 d half-life of 88-Zr is convenient for this purpose. The aim of the present study was to investigate the production possibility of 88-Zr using medium-energy proton accelerators. Our recent systematic studies provided reliable cross-section databases both for the (p,2n) reaction on natural Y and the (p,X) nuclear process on natural Zr. This revealed that the former reaction can be very useful for production purposes below 40 MeV while the latter is the nuclear process of choice above 30 MeV. Based on the evaluated cross-section databases, the integral 88-Zr yields for natural yttrium(III) oxide and elemental Zr targets have been calculated as a function of energy. This enabled the optimization of the 88-Zr production yield for a practical tandem-target arrangement. In the case of a 66 MeV incident proton beam, the available optimal energy windows are 62.6 MeV down to 35.6 MeV for metallic Zr and 32.2 MeV down to threshold for yttrium(III) oxide, providing a total physical yield just above 21 MBq/uAh. Note that such beams are routinely available at iThemba LABS for radionuclide production purposes. A similar study for a 70 MeV incident beam was performed in view of the new generation of commercial 70 MeV cyclotrons now available on the market. In this case, the optimal energy windows are 66.7 MeV down to 35.8 MeV for metallic Zr and 32.4 MeV down to threshold for yttrium(III) oxide, providing a total physical yield of 23 MBq/uAh. A generator is retained as a stock solution and only loaded onto a freshly prepared ion-exchange resin column prior to the separation of the 88-Y. Columns are used only once and used columns are discarded as waste.

Log: 144. **SPECTROSCOPIC SIGNATURES FOR FORENSIC SCIENCES.** Wozniak, N.R. (1,2); Barefield, J.E. (2); Clegg, S.M. (2); Czerwinski, K.R. (1); Judge, E. (2); Wagner, G.L. (2); Wilkerson, M.P. (2). (1) University of Nevada, Las Vegas. (2) Los Alamos National Laboratory.

Signatures arising from production, conversion, and aging processes are chemical in nature, and optical measurements reveal the potential to detect persistent molecular signatures characteristic of material origin, age or process history. The ability to quantitatively identify new anthropogenic signatures from natural background signals is an important theme for forensic sciences. The characterization of vibrational structure of uranyl materials and minerals formed under environmental conditions will be used to evaluate spectra collected from anthropogenic uranium bearing materials and uranium minerals. Vibrational structure will be determined using Infrared and Raman Spectroscopies. Anthropogenic uranium – bearing materials will include uranium compounds



commonly found throughout the mining and processing of uranium ore, fabrication of uranium fuel from uranium ore concentrates, and the reprocessing of spent uranium fuel, as well as, uranium bearing minerals. Here, we describe efforts in material synthesis and characterization of infrared absorption and Raman spectra and the instrumentation used to measure these signatures. LA-UR-14-28855

Log: 145. **A RADON AMPLIFIER FOR ATMOSPHERIC MEASUREMENT.** Topin, S. (1); Richon, P. (1). (1) CEA/DAM Ile de France.

The monitoring of naturally occurring radionuclides (e.g.  $^7\text{Be}$ ,  $^{10}\text{Be}$ ,  $^{210}\text{Pb}$ , and  $^{222}\text{Rn}$ ) is considered as an essential component of the WMO (World Meteorological Organization) for studying atmospheric processes. Regarding its short half-life (3.82 days) and its poor solubility/reactivity, the radon 222 is a key gas for environmental monitoring. It could be used in a many ways: to monitor the air mass movement, for back trajectory computation to identify the air mass origin, to trace recent (<3 weeks) air-mass contact with ice-free terrestrial sources. Nevertheless, due to the low radon gas activity, it could be hard to measure the radon 222 activities, especially at a high frequency (< one hour), with the usual detectors: alpha spectrometry, ionization chamber, scintillation, PIPs detectors, etc. The CEA developed a radon 222 amplifier, based on the use of a compressor and permeation membranes, that allows to enhance the radon 222 concentration by at least 70. When coupling with a commercial radon 222 gas detector as the AlphaGUARD™ ionization chamber, this innovative process enables to reach a unprecedented detection limits of about 40 mBq m<sup>-3</sup> for a 1 h measurement, of about 3 Bq m<sup>-3</sup> for a 10 min measurement and of about 14 Bq m<sup>-3</sup> for a 1 min measurement.

Log: 146. **COMBINED PHOTON / NEUTRON RADIOGRAPHY FOR NONDESTRUCTIVE ANALYSIS OF MATERIALS.** Barzilov, A. (1); Hartman, J. (1). (1) University of Nevada Las Vegas.

The nondestructive evaluation of complex objects including their shape and material composition is important to many applications in industry, security, and medicine. The feasibility of combination of photon and neutron radiography for nondestructive analysis and characterization of materials was studied. The MCNP6 code was utilized to model radiation transport processes in a dual-radiation tomography system. The computational model was composed of a fusion neutron source, a bremsstrahlung photon source, and a radiation detector array. An object under scrutiny was positioned between the source and the detector. The objects used in the study consisted of a matrix of low-Z and high-Z materials of various shapes and density. Transmission-radiography computations were carried out for fan beams with linear detector geometry. The tallies for both neutron and photon flux were modeled for the same geometry of the system. The ratios of the photon- to neutron-transmission were computed for pixels of the detector array. Using the inherent difference between neutron and photon interactions with nuclei of the interrogated object, it was possible to determine the shape and material composition of complex samples. It was shown that combined neutron / photon radiography allows better characterization of materials compared to the dual-energy photon radiography. The use of an array of scintillation detectors for simultaneous measurements of fast neutrons and photons is discussed for application in the neutron/photon radiography.

Log: 147. **ADVANCES IN AGE DATING OF NUCLEAR MATERIALS FOR NUCLEAR FORENSIC PURPOSES.** Zsolt Varga, Adrian Nicholl, Maria Wallenius, Klaus Mayer.

Age dating of nuclear materials (uranium and plutonium) is one of the most important signatures to assess the possible source of an unknown illicit material [1]. The principle derives from the presence of the trace-level decay product of radioactive nuclides, thus ideally based on the parent to daughter ratio the production date of the material (age) can be measured. The aim of the present study is to show the recent developments in uranium and plutonium age dating. A novel methodology was proposed to prepare a uranium age dating reference material based on the  $^{230}\text{Th}/^{234}\text{U}$  chronometer [2]. The principle was validated by a joint co-operation of expert nuclear forensic laboratories worldwide. The study aimed to compile the best practices, and also demonstrated the current limitations and state-of-the-practice. Based on the methodology a unique certified uranium age dating reference material was prepared in a joint undertaking by the Institute for Transuranium Elements and the Institute for Reference Materials and Measurements of the European Commission Joint Research Centre. The prepared certified reference material (candidate IRMM-1000) will help nuclear forensic laboratories to improve their nuclear forensic capabilities, as well as it can support basic research, such as half-life re-determination, in order to improve precision and accuracy [3]. 1. Mayer K., Wallenius M, Varga Z (2013), Chemical Reviews 113, pp884 2. Varga Z, Nicholl A, Wallenius M, Mayer K (2012), Analytica Chimica Acta 718, pp25 3. Varga Z, Nicholl A, Mayer K (2014), Physical Review C 89, pp064310

Log: 148. **SOURCE APPORTIONMENT OF FINE AIRBORNE PARTICLES IN XINZHEN,BEIJING USING POSITIVE MATRIX FACTORIZATION.** Jin Xiangchun(1);Zhang Guiying(1);Xiao Caijin(1);Huang Donghui(2);Yuan Guojun(1);Yao Yonggang(1);Wang Xinghua(1);Hualong(1);Wang Pingsheng(1);Ni Bangfa(1).(1)China Institute of Atomic Energy.(2)Ministry of Environmental Protection.

A total of 140 airborne particulate matter samples were collected at Xinzhen from May 19,2007 to July 19,2013 and their chemical composition were analyzed by Particle Induced X-ray Emission (PIXE) and Energy Disperse X-ray Fluorescence (ED-XRF). The possible sources of PM<sub>2.5</sub> were identified by positive matrix factorization (PMF). Five major sources have been identified and their corresponding apportionments estimated in Xinzhen. They are coal burning(29.2%), vehicle exhaust and waste incineration(26.3%), construction industry(23.3%), soil(15.4%) and industry with chlorine(5.9%). The trajectories of sources have been analyzed using sources contribution rates and global meteorological data. Conditional probability function (CPF) and the potential source contribution function (PSCF) were used to determine where sources came from. Extreme events, such as sandstorms, were identified and traced.

Log: 149. **AN ANALYTICAL SOLUTION OF NON-LINEAR EQUATION OF CHROMATOGRAPHY AND ITS APPLICATION IN DETERMINATION OF RADIOACTIVE STRONTIUM.** Grahek, Ž.; Coħa, I. Ruđer Bošković Institute.

The ideal linear model of chromatography can be used for the modelling of radionuclide sensor column in so called "equilibration based pre-concentration mini-column sensor analysis for the determination of low level concentration of some elements and radionuclides in water"[1]. Egorov et al. used theoretical approach typical for linear equilibrium chromatography and applied theoretical solutions for frontal analysis to modelling (predict) column-detector response (column with low number of theoretical plates - solutions for the breakthrough curves). This approach is reasonable because at low concentration of element and "short" column, linear sorption isotherm and low number of theoretical plates are expected. However, if sample contains higher concentration of target element, deviation from linear condition is expected. In open literature, simple analytical solution doesn't exist for non-linear isotherm in case of frontal analysis. Therefore, in this paper authors will show how an analytical solution for parabolic isotherm can be obtained by combination of Cole-Hopf and Laplace transform. This solution will be used to predict sensor column response in previously mentioned sense. Difference between linear and non-linear solution with emphasis on prediction of Sr binding profile on Sr specific reins (Sr resin, SuperLig) and expected column response in case of low number of theoretical plates will be discussed. The advantages and limitations of non-linear solution will be shown through application to determine radioactive strontium in real samples.

Log: 150. **ON COLUMN CHERENKOV DETECTION OF SR-89,90.** Coħa, I.; Grahek, Ž. Division for Marine and Environmental Research, Ruđer Bošković Institute.

In recent years fully automated approach for monitoring pure beta emitters have been developed [1-5]. In our laboratory semi-automated procedure for determination of Sr-89,90 in water samples was established [6]. Procedure includes separation of strontium on Super Lig 620, from matrix followed by off line on column detection via Cherenkov counting, on commercially available instrument, LSC Tri-Carb 3180 TR/SL. The goal is further development of mentioned method for monitoring environmental occurrence of Sr-90. The major influence on detection limit have separation and preconcentration on the column as well as detection efficiency. Therefore, optimal conditions for Cherenkov on column detection will be established by varying type of column and vial material, thickness of column walls as well as media around the column. The influence of mentioned parameters on Cherenkov counting efficiency of Sr-89,90 will be discussed.

Log: 151. **COMPARISON OF NUCLEAR FUEL COMPOSITION IN COMMERCIAL NUCLEAR FUEL CYCLE WITH GAMMA-RAY SPECTROSCOPY.** Egnatuk, C.M. (1); Wang, T.F. (1). (1) Lawrence Livermore National Laboratory.

ORIGEN-ARP was used to simulate the commercial nuclear fuel cycle for both uranium oxide fuel and mixed oxide fuel in a light water reactor. Fuel with varying burn-ups was used to create a picture of the isotopes composition and activity. MCNP6 was used to simulate high-purity germanium gamma-ray spectra for different snapshots in time of the fuel cycle. Gamma-ray intensity ratios of fission products in the mass chains 89 through 92 and 131 through 141 were used to distinguish the fuel type. These ratios were also compared to the gamma-ray intensity ratios from fission of a single isotope, i.e. U-235, Pu-239 or U-238, to create a method of differentiating the source of the fission.

Log: 152. **Novel Radiometric Age Determination for Uranium Material.** Bowers, D.L. (1), Orlandini, K.A. (1), Croatto, P. (2). (1) Argonne National Laboratory. (2) New Brunswick Laboratory.

New methods to estimate the age of uranium bearing materials were developed, employing daughter chain isotopes. Both primary and dual chronometers (e.g., parent/daughter and daughter/daughter isotopic ratios) were investigated. The age of the uranium bearing material, such as low enriched UF<sub>6</sub>, low enriched UO<sub>2</sub>, high enriched U<sub>3</sub>O<sub>8</sub> and uranium ore, is defined as the last date of the uranium purification. The alternative methodology uses isotopic ratio chronometers that require no isotopic dilution (ID) or the addition of radiometric tracer addition(s). Comparisons were made to the standard ID method for known reference materials. The results of these dual radiometric chronometers ("clocks") show no bias and are within the estimated measurement errors.

Log: 153. **RADIONUCLIDE ANALYSIS OF VARIOUS TYPES OF WATERS AND SEA SEDIMENTS FROM FUKUSHIMA.** Steinhäuser, G.(1); Shozugawa, K.(2), Riebe, B. (3). (1) Colorado State University. (2) University of Tokyo. (3) University of Hannover.

In this study we investigated various samples from Japan for possible radionuclide contaminations from the Fukushima nuclear accident. Target radionuclides were Cs-134 and -137, Sr-90, H-3, and I-129. Sample types included sea sediments off the coast of Fukushima prefecture, ocean water, tap water and one sample of reactor coolant from the Fukushima Daiichi reactor unit #5. Levels in water were generally low, however, in tap water, radiocesium could be detected. Due to very limited sample volume, in contrast to ocean sediments, no anthropogenic radioactivity could be detected in ocean water. The most remarkable result is that the reactor coolant did not exhibit detectable amounts of any other radionuclide than tritium. Even the search for increased I-129 using accelerator mass spectrometry revealed very low levels of this radionuclide. This is impressive evidence for the performance of the purification of the reactor coolant.

Log: 154. **OUR PET PROJECT: AN UNLIMITED SUPPLY OF BIG AND SMALL WATER SAMPLE VIALS FOR THE ASSAY OF RADON IN WATER.** Lane-Smith, Derek (1); Tuccimei, Paola (2); Galli, Gianfranco (3); Simko, Jesse (1); Cook, Ian (1); Bond, Charlotte Emily (1); Lucchetti, Carlo (2); De Simone, Gabriele (2). (1) DURRIDGE Company, Billerica MA, USA, (2) "Roma Tre" University, Dipartimento di Scienze, Roma, Italy, (3) Istituto Nazionale di Geofisica e Vulcanologia, Sezione Roma, Roma, Italy.

While the RAD AQUA accessory of the RAD7 radon detector provides for continuous, highly sensitive radon monitoring in open water and the RAD H2O is designed for the analysis of discrete ground water samples, a system to make good measurements of samples from both environments has been sought for some time. Big bottle systems have been developed, using large, glass bottles, or jugs. But these are fragile, and may be too large for groundwater measurement. In searching for a better solution, it was noted that Coke and similar bottles, made of PET (Polyethylene terephthalate), are engineered to contain CO<sub>2</sub> at the high pressures they may experience if heated by the sun. Experiments, consisting of taking multiple samples of a steady water source and analyzing them on different days, determined that radon leakage from a water sample contained in such a bottle was small compared with the loss by radioactive decay. Aeration adaptors have been manufactured to fit these bottles and CAPTURE software has been upgraded, with graphics and analysis, for every size of Coke bottle. A theoretical analysis and experimental verification of the system will be presented. Plastic Coke and similar bottles, made of PET, provide a global and virtually unlimited supply of water sample vials ranging in size from 350 ml to 2L or more. The RAD7, with the Soda Bottle Analysis System and CAPTURE software, provides an automatic aeration and analysis system suited to all sizes of Coke bottle and can measure radon concentrations in the water from 1 to 60,000 pCi/L.

Log: 155. **Removal of Tc from Neutron-Capture Mo-99 using Eichrom's ABEC Resin.** Bennett, M.E. (1); De Vries, D. (2); Isensee, G. (2); Beebe, D. (2); Harvey, J. (2); Vandegrift, G.F. (1). (1) Argonne National Laboratory (2) NorthStar Medical RadioIsotopes.

NorthStar Medical Radioisotopes is pursuing a neutron capture [Mo-98(n,γ)Mo-99] route for Mo-99 production at the University of Missouri Research Reactor (MURR). Argonne is assisting NorthStar in the development of some aspects of the operation. Once the molybdenum targets are removed from the reactor, they will undergo dissolution. The output of the dissolution results in ~1500 mL of a 5 M KOH solution containing ~400 Ci of Mo-99, as well as a few byproducts (compared to fission) including Tc. This solution is pumped from a shielded cask through a chromatography column containing ABEC to remove Tc present from the target irradiation. This polishing step will allow radiopharmacies to use the first aliquot of Tc they elute from their generators. Currently, radiopharmacies discard the first aliquot from the generator, as it contains unacceptable levels of Tc-99g. An ABEC cartridge has been tested according to the FDA PAT study requirements. The development of the ABEC cartridge and the PAT study results will be discussed.

Log: 156. **A NOVEL NEW BENCH TOP APPROACH TO THE ISOTOPIC PURIFICATION OF PU244 AND ITS APPLICATIONS.** Liezers M. (1); Farmer O.T. (1); Thomas M.L. (1); Hager G.J. (1); Eiden G.C. (1). (1) Pacific Northwest National Laboratory.

A new bench top approach to isotopic purification has been developed and applied to the production of a small quantity of Pu244 with an isotopic purity >99.99%, as compared to the standard Pu244 available that displays an isotopic purity of 97.87%. The presence of the Pu isotopes 238, 239, 240, 241 and 242 have been greatly reduced, allowing for higher spiking levels of the isotopically purified Pu244 tracer that should improve trace and ultra-trace level Pu measurements. Details of the isotopic purification approach will be described along with the reduction factors achieved for the Pu isotopes 239, 240, 241 and 242 and the effect this improved Pu tracer can have on analytical Pu mass spectrometry measurements.

Log: 157. **GEOCHRONOLOGY OF PUGET SOUND SEDIMENT USING SI-32: REDUCTION IN BACKGROUND RADIOGENIC ISOTOPES FOR COUNTING ON AN ULTRA-LOW-BACKGROUND BETA DETECTOR.** Finch ZS, BN Seiner, LM Arrigo, JE Strivens, ME Keillor, TW Hossbach, and GA Gill. Pacific Northwest National Laboratory.

This project is focused on developing a geochronology tool enabling age dating of coastal marine sediments in the 100-1000 year age range. The technique employs the  $^{32}\text{Si}/^{32}\text{P}$  radio-chronometer with an ultra-low-background gas proportional beta detector. This detector is anticipated to have a background count rate approaching 10 counts per day (cpd), thus requires significant decontamination of radiogenic nuclides present in the original sediment samples. This paper describes the  $^{32}\text{P}$  separation and purification procedures and the radiogenic background reduction needed for  $^{32}\text{Si}$  age-dating determinations. Sediment samples were processed through a heavy-liquid polytungstate separation to enrich the biogenic silica content, followed by an alkaline leach to extract the biogenic silica and associated  $^{32}\text{Si}$ . The silica was purified and then separated from the  $^{32}\text{P}$ . The  $^{32}\text{P}$  was further purified using multiple precipitation reactions. Aliquots from each chemical processing step were analyzed by an ultra-low-background 14 crystal germanium detector array and the associated chemical yields for Si and P were assessed by Inductively Coupled Plasma Optical Emission Spectrometry. The primary radiogenic contaminants found in the raw sediment were  $^{40}\text{K}$ ,  $^{137}\text{Cs}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ac}$  and had a total activity of approximately 18 Bq/g with the largest contributor being  $^{40}\text{K}$  with an estimated contribution of 83% of the total activity. In the final purified  $^{32}\text{P}$  sample a weak signal in the gamma spectra remained for thorium decay chain isotopes ( $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$ , and  $^{208}\text{Tl}$ ); however, levels were below the quantification limit for all nuclides. Further details on the purification and level of decontamination achieved are presented in this paper.

Log: 158. **RED OIL IDENTIFICATION, PRODUCTION CONDITIONS, AND MITIGATION STRATEGIES.** Huckabay, H.A.(1); Lee, D.L.(1); Birdwell, J.F.(1); Cable-Dunlap, P.R.(1). (1) Oak Ridge National Laboratory.

Red oil is a widely known potentially unstable mixture that is produced in spent nuclear fuel reprocessing facilities when a mixture of uranyl nitrate, nitric acid, tributyl phosphate (TBP), and a hydrocarbon diluent is heated. Explosive incidents involving this mixture have occurred at reprocessing facilities located in Savannah River, Hanford, and Oak Ridge when the organic phase (TBP and diluent) unintentionally entered an evaporator or calciner. Red oil production is an ongoing concern in reprocessing facilities, as it is widely believed that it can be formed at elevated temperatures. Recent work, however, has shown evidence that this mixture can also be formed under ambient conditions, which could impact long-term storage options for waste solutions. This work involves the production of red oil using known conditions to determine its gaseous composition; these results are compared to results from similar mixtures stored at ambient conditions. Several diluents are examined, and the gaseous effluents produced during heated conditions are characterized via gas chromatography-mass spectrometry. Based on these results, several recommendations will be presented to mitigate red oil production and improve long-term storage options for waste solutions in nuclear reprocessing facilities.



Log: 159. **STABILITY EVALUATION AND CORRECTION OF A PULSED NEUTRON GENERATOR PROMPT GAMMA ACTIVATION ANALYSIS SYSTEM.** Shypailo RJ, USDA/ARS Children's Nutrition Research Center, Department of Pediatrics, Baylor College of Medicine, Houston, TX 77030.

In prompt gamma neutron activation, the stability of source output is important for accurate quantification of gamma signals, especially for signals coming from low-concentration elements. One such element of interest, nitrogen, comprises only about 2.5% by weight of the human body. Assessment of total body nitrogen (TBN) can be used to estimate total body protein, thus it is an important body composition component at the molecular level. A system using a compact DT neutron generator ( $\sim 10^9$  n/s) has been developed for the measurement of TBN in vivo. Data acquisition is done using list mode, which allows for the assessment of event accumulation over time. Two whole-body anthropomorphic phantoms – one containing a nitrogen solution and one filled with water – were measured ten times each to assess instrument precision. Neutron generator stability was evaluated indirectly by reviewing patterns of event production over time. In addition, an auxiliary detector positioned away from the primary target captured total event production during measurements. Both the event/time patterns and the auxiliary detector showed significant variations among the repeated phantom runs. Neutron generator instability had a significant effect on TBN measurement precision. Measurement results were then adjusted based on the simultaneously acquired data from the auxiliary detector, which reduced the differences between phantom results and improved measurement precision. The DT neutron generator used in our system must be monitored for consistency of output, and adjustments must be made to measurement results to correct for generator instability.

Log: 160. **COMPARING FLUID ADVECTION RATES BETWEEN NATURAL OIL AND BUBBLE SEEPS IN DEEP OCEAN SYSTEMS USING RADIUM ISOTOPES.**

Peterson, L. (1); Peterson, R. (1); Joye, S. (2); Meile, C. (2); Montoya, J. (3); Webber, S. (3). (1) Coastal Carolina University, School of Coastal and Marine Systems Science, Conway, SC 29526. (2) University of Georgia, Department of Marine Sciences, Athens, GA 30602. (3) 3. Georgia Institute of Technology, School of Biology, Atlanta, GA 30332.

Natural hydrocarbon (e.g., oil and methane) seeps along the ocean floor supply the overlying water with diffuse carbon and nutrient loads. These deeply-sourced fluid seeps provide critical biogeochemical constituents supporting chemosynthetic communities around the discharge features, yet our understanding of the rate of fluid and material supply relative to the nature of the discharging fluid is limited by our current methods to constrain flux estimates. Remote sensing techniques used to quantify hydrocarbon discharge offer large-scale estimates without characterizing small-scale seepage variability that is important in shaping the benthic and pelagic ecosystem response. Here, we present a novel method coupling in-situ porefluid profile  $^{224}\text{Ra}$  observations with laboratory  $^{224}\text{Ra}$  ingrowth experiments from several oil and methane rich Gulf of Mexico benthic environments. Using a natural, conservative radiotracer with a 3.6 day half-life, we constrain in-situ production and decay rates of the porewater entrained radium, for each layer in a given profile for use in a 1-dimensional advection-diffusion equation to generate surficial fluid flux estimates to overlying waters. Results indicate advection rates on the order of tens to hundreds of centimeters per minute with a high degree of spatial heterogeneity both within sites and among sites (e.g. oil versus methane seeps). This new approach using Ra to quantify exchange rates between the sediment-water interface lends itself to determining material fluxes in any fine-grained advective environment.

Log: 161. **TRANSITION-EDGE-SENSOR MICROCALORIMETERS FOR TRACE ACTINIDE ANALYSIS.** A.S. Hoover (1); E.M. Bond (1); M.P. Croce (1); G.J. Kunde (1); M.W. Rabin (1); L.E. Wolfsberg (1); D.A. Bennett (2); J. Hays-Wehle (2); D.R. Schmidt (2); D. Swetz (2); J.N. Ullom (2). (1) Los Alamos National Laboratory, Los Alamos, NM. (3) National Institute of Standards and Technology, Boulder, CO.

A new category of sensor is being developed for analysis of trace quantities of actinides with advantages over conventional alpha-spectroscopy and mass-spectrometry methods. We describe the application of superconducting transition-edge-sensor (TES) microcalorimeter detectors to the measurement of the Pu-240/Pu-239 mass ratio and the determination of Pu material age from picogram-quantity samples. By embedding the sample material inside a 4-pi radiation absorber, we measure the total reaction energy (Q-value) for each nuclear decay, producing a single spectral peak for alpha-decaying isotopes. This removes peak overlaps, resulting in a simpler spectral analysis problem. Using a low-noise TES detector, energy resolution of 1 keV FWHM has been achieved, an order of magnitude better than for conventional alpha-spectroscopy. We also present a new method for measuring Pu material age using the Pu-241/Am-241 decay chain, where both isotopes are measured simultaneously to determine the isotope ratio. The sample preparation method is simple and uses droplet evaporation on a gold foil with subsequent mechanical kneading of the absorber material, avoiding radiochemical separations or electrodeposition commonly used for mass-spectrometry and alpha-spectrometry measurements. We present quantitative measurements of Pu-240/Pu-239 ratio and Pu material age from aqueous solution samples using a TES microcalorimeter, and compare to known values. We expect the method can ultimately be applied to the analysis of single particles.

Log: 162. **IMPROVED SENSITIVITY OF LIQUID SCINTILLATION COUNTING FOR MEASUREMENT OF ENVIRONMENTAL RADIOISOTOPES.** Douglas, M.(1); Bernacki, B.E.(1); Erchinger, J.L.(1,2); Finn, E.C.(1); Keillor, M.E.(1); Morley, S.M.(1); Orrell, J.L.(1); Panisko, M.E.(1); Warren, G.A.(1); Wright, M.E.(1). (1) Pacific Northwest National Laboratory. (2) Texas A&M University.

Liquid scintillation counting (LSC) is a versatile and commonplace method for measurement of charged particle emitting radionuclides. LSC provides utility in a range of environmental science applications including hydrological studies of water transport, anthropogenic releases of radionuclides to the environment, and assessing vertical mixing rates within oceans. Instrumental measurement background is one limiting factor of radiometric measurement sensitivity. As part of the development of a custom low background LSC system located in a shallow underground laboratory at Pacific Northwest National Laboratory, a number of measurement applications of LSC have been considered and are summarized here. The focus is on determining which aspects of such measurements would gain the greatest benefit from the reduction of LSC backgrounds by a factor of 10-100 relative to values reported in the literature. Examples of benefits include lowering the minimum detectable activity, reducing the sample size required, and shortening the elapsed timeline of the processing and analysis sequence. In particular tritium, strontium, and actinium isotopes are examined as these isotopes cover a range of requirements related to the LSC measurement method (e.g., tritium: low energy beta emission; radiostrontium: beta spectral deconvolution; actinium: alpha/beta discrimination). In this paper we summarize reported measurements of these isotopes in environmental science applications and illustrate the impact a low-background LSC system can be expected to provide.

Log: 163. **PLUTONIUM ISOTOPES IN SOILS FROM THE DOWNWIND AREA OF LOP NOR NUCLEAR TEST SITE: REGIONAL FALLOUT VS GLOBAL FALLOUT.** Bu, W. T. (1,2); Guo, Q.J. (1); Zheng, J. (2); Uchida, S. (2). (1) School of Physics, Peking University. (2) Research Center of Radiation Protection, National Institute of Radiological Sciences.

Pu isotopes in the downwind areas of nuclear test sites should be investigated for radioactive source identification and radiological assessment. For the first time, soil core samples from Jiuquan region in the downwind area of Lop Nor nuclear test site (CNTs) were analyzed for Pu isotopes. The  $^{239+240}\text{Pu}$  activities and  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios were obtained. The high Pu inventories in most of the sampling places revealed that this region was contaminated by the regional fallout Pu from the CNTs. The deposition of Pu from the CNTs in the downwind area was heterogeneous as the Pu inventories showed large variations. The contributions of the CNTs to the total Pu in the soils were estimated. The averaged  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio for all the soil samples was around 0.16, slightly lower than that of global fallout. This atom ratio could be considered as a mixed fingerprint value representing Pu isotopes from the CNTs. Pu in the soils of Jiuquan region showed faster downward migration rate compared with other investigated places in China.

Log: 164. **EVALUATION OF ENVIRONMENTAL RADIATIONS AT AXESSE LINAC OF MEDICAL UNIVERSITY UNDERGOING TREATMENT OF VOLUMETRIC-MODULATED ARC THERAPY (VMAT).** Tseng, H.C. (1); Liu, W.S.(4); Lai, W.H.(3); Huang, H.S.(3); Lin, J.B.(2); Chen, C.Y.(3). (1) Department of Radiation Oncology, (2) Department of Medical Image, Chung Shan Medical University Hospital, (3) School of Medical Imaging and Radiological Science, Chung Shan Medical University, (4) Department of Radiation Oncology, Kaohsiung Veterans General Hospital.

A novel radiation technique, volumetric-modulated arc therapy (VMAT) of Axesse (Elekta 2538) linear accelerator (linac) has been introduced because of important treatment for lung cancer. So far, no study with environmental radiations for treatment room was published. This work is long-term monitoring environmental radiations using Thermoluminescent (TL) dosimeter at linac of the medical university. First, optimizing the conditions of TL dosimetry reading system, the reading system was optimized for initial temperature, heat rate, and maximum set temperature using the Taguchi methods. The major algorithm is built according to the Taguchi analysis and tries to apply onto the values of environmental radiation of treatment room. The optimized values are (1) 50°C for initial temperature, (2) 10 °C/s for heating rate (3) 240°C for the maximum temperature. Environmental radiation of up to 3.66 Sv/mo at ceiling of the gantry head indicated heavy scattering photon spread into treatment room. Two dimensional distributions of photo radiations had been mapped. In addition, the half value layer of 6 MV photons was evaluated. The minimum detectable dose using this method was also discussed. The quantitative results could provide practical guidance regarding radiation protection to workers, patients, the relatives and the public.

Log: 165. **ON THE FORMATION OF NON-RADIOACTIVE COPPER DURING THE PRODUCTION OF  $^{64}\text{Cu}$  VIA PROTON AND DEUTERON INDUCED NUCLEAR REACTIONS ON ENRICHED  $^{64}\text{Ni}$  TARGET.** Szelecsényi, F.(1); Steyn, G.F.(2); Kovács, Z.(1). (1) ATOMKI, Debrecen, Hungary. (2) iThemba LABS, Faure, South-Africa.

The production of radioisotopes in high specific activity form, especially in the case of the majority of PET nuclides, is a continuous challenge for many laboratories. The radioisotope products usually could be contaminated not only with other radioisotopes but with different stable elements as well. With properly selected targetry, irradiation circumstances, preparation and separation methods, however both the radio-nuclide and stable element contaminations can be decreased to acceptable levels. Unfortunately in the case of production of  $^{64}\text{Cu}$  (PET isotope, half-life = 12.7 h; formed via:  $^{64}\text{Ni}(p,n)$  or  $^{64}\text{Ni}(d,2n)$  nuclear reactions), non-radioactive copper ( $^{63}\text{Cu}$  and/or  $^{65}\text{Cu}$ ) are also co-formed (above 10 and 3 MeV, respectively). Since this 'cold copper' contamination remains un-separable in the final product, its presence could decrease dramatically the specific activities of any copper-labeled radiopharmaceuticals. Based on the excitation functions for the  $^{64}\text{Ni}(p,2n)^{63}\text{Cu}$ ,  $^{64}\text{Ni}(d,n)^{65}\text{Cu}$  and  $^{64}\text{Ni}(d,3n)^{63}\text{Cu}$  reactions, (estimated via the TALYS and ALCE2014 codes), in this work we calculated the ratio of nat-Cu/ $^{64}\text{Cu}$  as a function of bombarding energies and irradiation time. It was found that increased production energy window and/or longer irradiation time results in increased cold copper contamination levels in both production cases. The results also revealed that  $^{64}\text{Cu}$  can be produced in no-carrier-added form in the case  $^{64}\text{Ni}+p$  reaction but only below 10 MeV. Using a usual PET cyclotron (up to 16 MeV protons) this cold copper contamination level at EOB still remains below 20 % (supposing 10 h irradiation time). On the other hand the 'deuteron way' produces cold copper ( $^{65}\text{Cu}$ ) and  $^{64}\text{Cu}$  at the same time in the investigated energy region. In addition to this, above 14 MeV the formation of  $^{63}\text{Cu}$  is also possible. The  $^{65}\text{Cu}/^{64}\text{Cu}$  ratio at EOB will be around 0.9 using 8 MeV deuterons for 10 h. According to the present study, the proton production way results  $^{64}\text{Cu}$  with higher specific activity than the recently suggested deuteron method.

Log: 166. **APPLICATION OF RA-228 AS A QUANTITATIVE TRACER OF SUBMARINE GROUNDWATER DISCHARGE OVER GLOBAL AND REGIONAL SCALES.** Kim, G.; Kwon, E.Y.; Cho, H.M.; Kim, J. Seoul National University.

Ra-228 (half life: 5.75 years) has been utilized as an excellent tracer of submarine groundwater discharge (SGD) for basin and regional scales. In this study, based on the compiled global Ra-228 data, we calculated the flux of Ra-228 to the global coastal ocean using an inverse model. Then, the SGD-driven Ra-228 flux is estimated by subtracting contributions from the other sources including wind-blown dust, diffusive fluxes from coastal sediments, and rivers. By applying the end-member concentrations of the Ra-228 in coastal groundwater, the SGD is estimated to be  $(12 \pm 3) \times 10^{13} \text{ m}^3 \text{ yr}^{-1}$  over the Atlantic (30%) and Indo-Pacific Oceans (70%). This magnitude is 3 to 4 times greater than the freshwater fluxes into the oceans by rivers. In addition, we compiled Ra-228 data in the northwestern marginal seas (East China Sea, Yellow Sea, South Sea of Korea, and the East (Japan) Sea) of the Pacific Ocean, which seems to be a hot spot of Ra-228 fluxes to the Pacific Ocean. On the basis of a mass balance model, Ra-228 flux through SGD is estimated to be an order of magnitude higher than that in other typical continental margins, perhaps due to the larger tidal pumping. Thus, we suggest that SGD is the dominant pathway for inputs of terrestrial materials (including anthropogenic sources) to the global ocean and is more important in the northwestern marginal seas where the loading of anthropogenic chemical materials are rapidly increasing.

Log: 167. **ESTIMATING THE RESIDENCE TIME OF SEAWATER IN A SUBTERRANEAN ESTUARY USING A RADON TRACER.** Oh, Y.H.; Kim, J.; Kim, G. Seoul National University.

The activity of Rn-222 in coastal groundwater was monitored every two hours continuously from September 2010 to May 2011 in Uljin, Korea. The monitoring station is ~4 m off the shoreline, where the average water depth is 1.5 m. The inlet for water intake is positioned ~70 cm deep inside the sandy/rocky seafloor. The activity of Rn-222 was measured using a radon-in-air monitor (RAD7, DurrIDGE Co.) connected to an air-water exchanger. The activities of Rn-222 showed large variations ranging from 2,300 to 45,000 Bq/m<sup>3</sup>, with some unusually high peaks. As the mean sea-level falls from September 2010 to January 2011, the activities of Rn-222 decreased by a factor of 3–4. By subtracting the contributions of Rn-222 from fresh groundwater to each sample, we obtained the residence time (ingrowth time of Rn-222 in the aquifer) of recirculating seawater in the monitoring location. The residence times of recirculating seawater were close to 5 days, with higher values at low tides. The largest fluctuation of residence times were observed during spring tides. We suggest that Rn-222 can be used as a powerful tracer of seawater residence times in subterranean estuaries, which are important zones for geochemical reactions.

Log: 168. **STABILITY OF THE ORCA BASIN BRINE INTERFACE DETERMINED USING RADIUM ISOTOPES.** Peterson, R.N.(1); Peterson, L.E.(1); Montoya, J.P.(2); Joye, S.B.(3). (1) Coastal Carolina University. (2) Georgia Institute of Technology. (3) University of Georgia.

The Orca Basin is an intraslope basin in the northern continental slope of the Gulf of Mexico, and contains a deep (up to 220 m) anoxic, hypersaline brine lake. The brine interface extends from ocean salinity (at a water depth of 2125 m) to a constant salinity of ~215 psu below a depth of 2250 m. This interface is considered to be among the most stable interfaces on the planet, and contains distinct zones of macrofaunal and microbial assemblages. The brine lake is supported by continued dissolution of exposed rock salt at an estimated rate of 0.5 million t/yr. Such dissolution of salt should serve as a source of radium isotopes, and given their range of half-lives, these isotopes can serve as unique proxies to the location and rate of salt dissolution into this basin. We have collected a series of radon and radium isotope profiles through this brine lake over the past 4 years, which allow us to assess the spatial and temporal stability of this interface. Throughout the brine lake, we observe strong enrichments in unsupported Ra-224, Rn-222, and Ra-226 in a ~10 m thick zone near the base of the brine interface, between 2232 m and 2242 m. The strong enrichment in unsupported Ra-224 in this layer must be supported by a continuous source, presumably the dissolution of exposed salt rock. Various degrees of isotopic enrichment throughout the lake provide an assessment of the rates of lateral and vertical dispersion of salt and associated chemical constituents.



Log: 169. **PREPARATION OF THIN GADOLINIUM SAMPLES VIA ELECTRODEPOSITION FOR EXCITATION FUNCTION STUDIES.** Kovács, Z.(1); Szelecsényi, F.(1). (1) ATOMKI, Debrecen, Hungary.

The difficulty in preparation of rare earth elements by electrodeposition is well known. For example, the maximum thickness of Gadolinium prepared this way was only 0.5 mg/cm<sup>2</sup> with acceptable mechanical stability. Unfortunately it is not thick enough for several cross-section studies. Our aim was to prepare 2-3 mg/cm<sup>2</sup> thick Gd layers in order to study Gd+p and Gd+d reactions using highly enriched (> 90%) <sup>152,155</sup>Gd target materials. The above reactions produce the medically relevant <sup>152,155</sup>Tb radioisotopes. For electrodeposition, we developed a very small volume cell in order to provide suitable Gd electrolyte concentration in case of the very expensive target material. The cell consists of a small volume (2.8 cm<sup>3</sup>) glass tube located between two Teflon holders. The lower one holds the Al cathode on a support. We used Pt ring as anode, and no stirring was applied during the electrolysis because of the small volume. However, the H<sub>2</sub> bubbles, developed in parallel with the Gd deposition during the process, maintained the homogenous concentration in the electrolyte. It was found that the composition of the electrolyte is the major factor for the quality of the deposited layer. It was also very important to keep the H<sup>+</sup> concentration as low as possible in order to prevent the intensive H<sub>2</sub> bubbling. After preliminary testing of different concentration of Gd and composition of the organic-water solvent, a 3 mg/ml Gd<sup>3+</sup> concentration was applied in 0.5 ml 0.01 M HCl and 19.5 ml absolute ethyl-alcohol mixture as electroplating solution. From this solution 2.5 ml was used for each electrodeposition. The optimal quality Gd layer was prepared at constant 25 mA current during the whole process. Lower current resulted in slow deposition with thinner layer, while the intensive H<sub>2</sub> development at higher currents resulted in bad quality surface. With the electroplating solution described above a typical deposition time was 50-55 minutes. We stopped the electroplating when the development of H<sub>2</sub> began to rise very quickly. Under the conditions described above we prepared smooth and adhesive Gd layers up to 3 mg/cm<sup>2</sup>. At repeated experiments the variation of the thickness was between 1.6 to 3 mg/cm<sup>2</sup>. These samples can withstand 18 MeV proton beam up to 100 nA irradiation without any damage.

Log: 170. **Examining the ionization of refractory materials including actinides for electromagnetic isotope separation.** J.J. Horkley, E. Wählin, K.P. Carney, J.K. Pfeiffer, A.T. Johnson, M.R. Finck, J.E Davies (1)Idaho National Laboratory

An ion source test stand manufactured by Beam Imaging Solutions Inc. and the stable isotope mass separator at Idaho National Laboratory (INL) are being used to support the evaluation of three ion source types to ionize refractory metals such as Zr, W, and Th. Eventually Pu and Np will be evaluated using the INL radioactive isotope mass separator. Several variations of a hot-cathode type source and a radio frequency (RF) source are being examined to determine their feasibility for use in electromagnetic separators. A third spark-type source is also being developed to ionize relatively volatile compounds such as metal-halides, -hydrides, and organometallics. To date, ionization of metallic Zr, W, and Th has been

demonstrated on the ion source test stand using the RF source. Initial Zr<sup>+</sup> ion beam currents using a Zr metal charge were approximately 400 nA through a 6.35 mm diameter aperture. W<sup>+</sup> and Th<sup>+</sup> ion beam currents were approximately 200 nA and 26 nA, respectively, through a 2 mm diameter aperture using a 2% (by weight) thoriated tungsten charge. This research supports isotope separation efforts to produce standards that are used for test, measurement, and calibration standards in nuclear forensics. Information attained thus far will be described.

Log: 171. **Reference standard characterization: an inter-laboratory assessment of highly enriched Ba-134 for use as a reference standard.** J.J. Horkley(1); K.P. Carney(1); R.R. Lewis(1); J.D. Sommers(1); J.P. Crow (1); C.A. Poole(1); T.C. Giglio(1); J.J. Giglio (1); R.D. Ball(1); J.L. Mann(2); J.L. Steeb(3). (1) Idaho National Laboratory (2) National Institute of Standards and Technology.

The isotope production group at Idaho National Laboratory has been producing high purity isotopes with abundances greater than 99% using an electromagnetic isotope separator. Ba-134, Sr-84, and Zr-96 are stable isotopes that are planned to be enriched above 99% abundance, and be certified as chemical reference standards for use as isotope dilution mass spectrometry internal standards ("spikes"). Isotopes from natural Ba and Sr starting materials have been electromagnetically separated, implanted on to Al, Cu, or Ti foils and harvested by leaching with high purity HNO<sub>3</sub> solutions. Ba or Sr concentrations range from 0.1 to 10 µg isotope per gram solution and contain up to 1mg per gram solution of Cu or Al when Cu or Al foils are used as collection substrates. Those solutions are chemically purified to remove dissolved collection substrate material by solvent extraction using 4,4'(5')-di-t-butylcyclohexano 18-crown-6 (Eichrom®) in toluene. Alternative targets made from high purity Ti were used that were inert to the acid leach and thus obviated the need for chemical purification. The dissolved isotopes were encapsulated in heat-sealed high purity quartz ampoules. In order to verify proper handling and sealing techniques for reference standard production, identical samples were packaged and sent to four laboratories for independent analysis. Initial results from the labs for Ba-134 solutions have shown consistent isotopic abundances (+/- 0.5%) in comparison to one another, demonstrating that the packaging and handling techniques are sound and that the introduction of contaminants can be minimized and controlled effectively. The results of the analysis of the Ba-134 samples by the multi-laboratory "round robin" will be presented.

Log: 172. **Ion beam characterization and collection methods to isolate low-abundance isotopes using electromagnetic separation.** J.E. Davies(1); J.J. Horkley(1); K.P. Carney(1; A.J. Edwards(1); J.J.Giglio(1); C.A. Zarzana(1) (1) Idaho National Laboratory.

Idaho National Laboratory currently uses a 90° sector electromagnetic isotope separator to produce high purity stable isotopes. A second separator of similar design is being optimized and will eventually be slated for the isolation of microgram quantities of actinide and other radioactive isotopes that can be used for a number of purposes including targets for cross section measurements and the production of reference standards for high precision mass spectrometry measurements. In order to effectively collect isotopes with near unity abundance, the ion beam should be well characterized and shaped to the collection system to provide efficient collection. In addition to beam shaping, a variety of collection substrates have been evaluated to improve collection efficiencies that are calculated based on recorded ion currents on the foil relative to the measured mass of each isotope that is determined by mass spectrometry. An acidic leaching solution was used to recover the product from each collection foil. For naturally abundant Ba isotopes that are collected in the non-radioactive mass separator, a 1 $\mu$ A beam current for any particular isotope would yield approximately 5  $\mu$ g hr<sup>-1</sup> of that isotope if all detected ions were recovered. Typical collection yields are less than the recorded ion current due to ion back scattering and spallation. In addition, the currents are often falsely high due to electron scattering on the foil, making the relative collected yield appear much lower. Efforts are ongoing to address these issues to maximize collection. In addition to collection studies, a multiple-Faraday cup detection array mounted on a linear slide is being developed and tested to create 3-dimensional images of measured ion beam profiles in the separator flight tube. These images can help determine the location of individual isotope focal points in the beam path and will assist in optimizing collection efficiencies for the new instrument.

Log: 173. **LOCATING AND IDENTIFYING TRACE PLUTONIUM PARTICLES ON HEPA FILTERS USING MICRO-X-RAY FLUORESCENCE.** Worley, C.G. (1); Tandon, L. (1); Martinez, P.T. (1); Decker, D.L. (1); Schwartz, D.S. (1). (1) Los Alamos National Laboratory.

Monitoring residue from plutonium operations is important for health and safety reasons. A particularly challenging application for micro-X-ray fluorescence (MXRF) here involved nondestructively locating plutonium particles trapped in high-efficiency particulate air (HEPA) filters for extraction and characterization. The HEPA filter samples analyzed contained a minimal amount of plutonium (~7 micrograms/cm<sup>2</sup> equivalent to ~50 pg of plutonium in the analysis beam). Although this would be too low to detect by MXRF, very isolated larger particles and aggregates were present dispersed over cm<sup>2</sup> areas. Nondestructively locating these particles directly on filters with Scanning Electron Microscopy (SEM) was impractical due to such large surface areas to scan with this high resolution technique, relatively poor Pu detection limits, particle and substrate charging, and potential radioactive contamination of the vacuum chamber. However, plutonium particles were successfully located using MXRF which does not have these limitations. Samples were analyzed through containment film to avoid instrument contamination. Locating the trace plutonium was extremely challenging due to severe energy overlaps with filter impurities, significant source scatter background, and ultra-low substrate particle densities. To overcome these challenges, instrumental parameters were optimized to locate plutonium particles that were then extracted with a micro-vacuum single stage impactor, collected on carbon tape, and morphologically and chemically characterized using SEM.

Log: 174. **LIQUID SAMPLING-ATMOSPHERIC PRESSURE GLOW DISCHARGE (LS-APGD) MICROPLASMA IONIZATION SOURCE: ELEMENTAL AND ISOTOPIC ANALYSIS IN THE LABORATORY AND MOVING TOWARD THE FIELD.** Marcus, R.K. (1); Zhang, L.X. (1); Harris, Jones, S.M.H. (1). (1) Clemson University.

Radiochemical and mass spectrometric (MS) methods are orthogonal in almost all practical aspects. Surely, the transportability of radiochemical instrumentation is one of its strong suites, while the benefits afforded by mass spectrometry are principally limited to a laboratory setting. Elemental/isotopic MS instrumentation is challenged in this regard by aspects involving the ionization sources and the mass analyzer platforms; we concentrate here on the former. Simply put, the inductively-coupled plasma (ICP) source requires too much operational overhead, and thermal ionization (TI) requires too much sample preparation to be applied outside of the laboratory. This laboratory is developing the liquid sampling-atmospheric pressure glow discharge (LS-APGD) microplasma for applications in elemental and isotopic analyses. This source operates at modest powers (<50 W, dc), with relatively low solution and support gas flow rates (10 uL and 10's mL per minute respectively). Beyond these favorable operation parameters, samples can be introduced into the microplasma as small (10 uL) solution aliquots, gases, or particulate aerosols produced via laser ablation. Initial source evaluation demonstrated mass sensitivities on the sub-nanogram level, with very high signal stability and a high level of robustness towards the introduction of samples of different matrix composition. Beyond elemental/isotopic analyses, the microplasma can be operated in a mode that also provides for determination of organics and identification of ligands appended to target lanthanides/actinides. We present an array of attributes that bode well for the further development of the LS-APGD as a pragmatic ionization source for field-deployable, elemental/isotopic MS analyses of natural and radioactive samples.

Log: 175. **EVALUATION OF A HYPER PURE GERMANIUM DETECTOR EFFICIENCY USING AN ANALYTICAL METHOD.** Abbas, M.I.(1); Faied, A.(1); Al-mugren, K.S.(2). (1) Physics department, Faculty of Science, Alexandria University, 21511 Alexandria, Egypt. (2) Physics department, Faculty of Science, Princess Nora Bint Abdul Rahman University, 11544-55532 Riyadh, Saudi Arabia.

A straightforward mathematical method for the efficiency calibration of HPGe detector is presented. The method is based on the model, developed earlier by Selim, Y.S. and Abbas, M.I. for gamma detector efficiency calculations. Furthermore, the full-energy peak efficiency (FEPE) and photofraction (P) of a HPGe detector were calculated and compared with the experimental measurements in the energy range between 60 and 1863 keV. The attenuation of photons by the source itself (self-absorption), the source container and the detector end cap materials is also included. By comparison, the calculated and experimental fepe and P values are in good agreement, the overall percentage error is less than 3%.

Log: 176. **SIMULTANEOUS REMOVAL OF VARIOUS IODINE SPECIES IN AQUEOUS SOLUTIONS OF HIGH SALT CONCENTRATIONS USING NOVEL FUNCTIONAL ADSORBENTS.** Tachibana, Y.(1); Tai, T.(1); Nogami, M.(2); Kaneshiki, T.(3); Nomura, M.(3); Suzuki, T.(1, 3). (1) Nagaoka University of Technology. (2) Kinki University. (3) Tokyo Institute of Technology.

We have been trying to find a new way out of effective removal of iodine species in contaminated water generated by means of sustained injections of seawater for emergency cooling of nuclear cores in Fukushima Dai-ichi Nuclear Power Station. Unfortunately, amounts of contaminated water with radionuclides such as iodine species has been increasing day by day and the total volume of the contaminated water has reached ca. 460,000 m<sup>3</sup>. As one of promising decontamination processes except for precipitation methods, it can be expected that adsorption separation processes by chromatographic techniques are available. However the removal capability of chromatographic processes using conventional adsorbents in aqueous solutions of high salt concentrations such as seawater has not been exactly accepted for environmental release. This difficulty of removal of iodine species is also attributable to various types of chemical forms of iodine species in aqueous solutions. In brief, insofar as we can tell, the available information on simultaneous removal of iodine species using effective adsorbents has not been reported yet. On the basis of the background described above, we have developed a novel hybrid-type adsorbent consisted of benzoimidazole combined with hydrolyzed tannic acid named BISTAS and adsorption mechanisms of iodine species using BISTAS in various types of original seawater and in various temperatures have been discussed in further detail, taking into account the results of DV-X $\alpha$  calculations of BISTAS.

Log: 177. **DETERMINATION OF ALUMINUM IN CANADIAN DUPLICATE DIETS BY A COMBINATION OF THERMAL AND EPITHERMAL INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS.** Sullivan, E.E.; Chatt, A. Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, 6274 Coburg Road, Room 212, P.O. Box 15000, Halifax, NS, B3H 4R2, Canada.

Exposure to aluminum is of public concern due to its potential toxic biological effects and association with Alzheimer's disease. Aluminum is one of the most common elements in the human environment. It is naturally present in water and most foods, and its levels vary with geographic location. Typical concentrations of aluminum in foods are 0.02–5 mg/kg (wet weight), and typical dietary intakes of aluminum are between 1 and 30 mg/d. Aluminum is a rather difficult element to determine by almost all analytical techniques. A combination of instrumental NAA (INAA) and epithermal INAA (EINAA) was applied in the present work using the Dalhousie university SLOWPOKE-2 reactor facility. The 1779-keV gamma-ray of  $^{28}\text{Al}$  (half-life=2.31 min) produced by the reaction  $^{27}\text{Al}(n,\gamma)^{28}\text{Al}$  was used for the determination of aluminum. EINAA was used to correct for contribution to the  $^{28}\text{Al}$  activity by the interfering reaction  $^{31}\text{P}(n,\alpha)^{28}\text{Al}$  from phosphorus present in the samples. Several reference materials were analyzed by the combination of the INAA/EINAA method and our results agreed well with the certified values. The methods were applied to 15 duplicate diet samples. The dietary intakes (dry weight, mg/d) for this population group had a mean of 7.0, median of 4.4, and the values ranged from 0.08–32.

Log: 178. **A COMPARISON OF EXPANDED UNCERTAINTIES FOR THE MEASUREMENT OF IODINE LEVELS IN FOODS BY PSEUDO-CYCLIC EPITHERMAL INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS WITH ANTICOINCIDENCE COUNTING USING SINGLE COMPARATOR AND K0 STANDARDIZATION PROCEDURES.** Nyarko, B.J.B. (1,2); Akaho, E.H.K. (2); Fletcher, J.J. (3); Chatt, A. (1). (1) Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, 6274 Coburg Road, Room 212, P.O. Box 15000, Halifax, NS, B3H 4R2, Canada. (2) Ghana Atomic Energy Commission, P.O. Box LG80, Legon-Accra, Ghana. (3) University of Development Studies, Tamale, Ghana.

We have developed several instrumental neutron activation analysis (INAA) methods for the determination of iodine in various foods from Ghana. These methods include conventional INAA, epithermal INAA (EINAA), pseudo-cyclic INAA (PC-INAA) and pseudo-cyclic EINAA (PC-EINAA). We have used conventional and anticoincidence (AC) gamma-ray spectrometry, and applied both single comparator and k0 standardization procedures in the above methods. We have evaluated the uncertainties associated with these methods. We have validated the methods by analyzing several reference materials (RM) and standard reference materials (SRM) obtained from the U.S. National Institute of Standards and Technology (NIST) and obtained good agreement with the certified values. Here we will present the expanded uncertainties associated with the measurement of iodine level in one of these materials, namely the NIST RM 8415 Whole Egg Powder, by a PC-EINAA-AC method using the single comparator and k0 standardization procedures. We obtained an expanded uncertainty ( $k = 2$ , ~95% CL) of  $1.81 \pm 0.48$  for the k0

procedure compared to that of  $1.85 \pm 0.22$  for the single comparator procedure. The value for the k0 procedure appears to be slightly influenced by the uncertainties in the nuclear constants and the detector full photopeak efficiency calibration.

Log: 179. **INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS COUPLED TO ANTICOINCIDENCE COUNTING SIGNIFICANTLY REDUCES INTERFERENCES FROM BROMINE AND ANTIMONY TO ALLOW RELIABLE MEASUREMENTS OF NANOGRAM LEVELS OF ARSENIC IN BIOLOGICAL MATERIALS.** Zhang, W. (1,2); Chatt, A. (1). (1) Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, 6274 Coburg Road, Room 212, P.O. Box 15000, Halifax, NS, B3H 4R2, Canada. (2) Radiation Protection Bureau of Health Canada, 775 Brookfield Road, AL 6302D1, Ottawa, ON, K1A 1C1, Canada.

Although instrumental neutron activation analysis (INAA) is a very sensitive technique for the determination of As, biological materials containing high levels of Br, K, and Na can produce high activities resulting in inferior detection limits of As. At sub-ppm or ppb levels of As commonly found in biological samples, the 559.1-keV peak of <sup>76</sup>As (half-life=25.9 h) is interfered with by the 554.3-keV peak of <sup>82</sup>Br (35.3 h) and 564.1-keV peak of <sup>122</sup>Sb (65.3 h). Modern Ge detectors usually have good enough resolution to resolve these three peaks. However, when Br and Sb levels are high, the 559.1-keV peak may be masked making its reliable measurement rather difficult, if not impossible. Anticoincidence counting (AC) technique should be ideal in such a situation. Concentrations of ppb levels of As in 16 RMs and SRMs containing various amounts of Br, Sb, K, Na and other possibly interfering elements were determined by INAA-AC. Background suppression ratio (BSR), defined here as the ratio of background counts under the 559.1-keV peak of <sup>76</sup>As in conventional spectrum to the background counts in the same region of the anticoincidence spectrum, varied significantly depending on concentrations of the major elements in the sample.



**Log: 180. MICELLE-MEDIATED EXTRACTION FOLLOWED BY NEUTRON ACTIVATION DETERMINATION OF LOW LEVELS OF ANTIMONY IN SEaweEDS.**

Serfor-Armah, Y. (1,2); Carboo, D. (3); Akuamoah, R.K. (3); Chatt, A. (1). (1) Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, 6274 Coburg Road, Room 212, P.O. Box 15000, Halifax, NS, B3H 4R2, Canada. (2) School of Nuclear and Allied Sciences, College of Basic and Applied Sciences, University of Ghana, P.O. Box AE 1, Atomic-Accra, Ghana. (3) Department of Chemistry, College of Basic and Applied Sciences, University of Ghana, P.O. Box 56, Legon-Accra, Ghana.

The use of micelle-mediated extraction, commonly known as cloud point extraction (CPE), is a good technique for preconcentration of trace elements. However, it has not been used that much in combination with NAA except in our and a few other laboratories. A simple one-step CPE method was developed for the preconcentration of Sb, considered a priority pollutant, using a mixture of PAN and TAN chelating agents and PONPE-20 surfactant. The parameters affecting the separation were optimized. The recovery of Sb under the optimum conditions of pH 6.4, [PAN/TAN]= $1 \times 10^{-4}$  M, [PONPE-20]=0.1% (m/v), ionic strength=0.05 M KNO<sub>3</sub>, and a temperature of 41°C was >95%. Antimony was assayed using the 564.1-keV photopeak of <sup>122</sup>Sb (half-life=2.72 d) after an irradiation for 3 h at a neutron flux of  $2.5 \times 10^{11}$  cm<sup>-2</sup> s<sup>-1</sup> in the Dalhousie University SLOWPOKE-2 reactor (DUSR) facility, decay for 7 d and counting for 15-20 h. The detection limit of the CPE method was 2.8 µg/kg in seaweeds. The method was validated using several reference materials. It was applied to the analysis of seaweeds.

**Log: 181. Neutron Spectrum Measurements from Irradiations at NCERC.**

Jackman, K. R.(1); Bredeweg, T. A. (1). (1) Los Alamos National Laboratory.

Monte Carlo simulations and radioactivation foil techniques were used to study the neutron spectra observed for COMET/ZEUS and Flatop assembly irradiations at the National Criticality Experiments Research Center (NCERC) at the Nevada National Security Site (NNSS). The neutron fluence spectra were modeled using the MCNP6 code. Activation foils included in these irradiations were analyzed using high-resolution gamma-ray spectrometry. The activation foil data was used with the SAND-II (SNL) and STAY'SL (PNNL) codes to unfold/adjust the neutron spectra. The unfolded/adjusted neutron spectra show reasonable agreement with the MCNP6 models.

Log: 182. **RAPID ACTIVATION PRODUCT ANALYSIS IN POST-DETONATION NUCLEAR FORENSICS.** Olson, A.C. (1); Kozimor, S.A. (1); Boland, K.S. (1); Bowen, S.M. (1); Oldham, W.J. (1); Schake, A.R. (1). (1) Los Alamos National Laboratory.

After the detonation of a nuclear device, obtaining rapid nuclear forensics conclusions is essential. One approach for determining the origin and type of nuclear device involves characterizing the isotopic ratios for products formed during the explosion. While several methods for diagnostic analyses center on fission product and actinide evaluations, quantification of activation products also offers the potential for providing useful information. However, activation product analyses have largely been ignored due to the technical challenges associated with their purification and analysis. Presented here is a unique approach for streamlining methods to purify, identify, and quantify activation product constituents of radioactive debris. We discuss the use of anion exchange chromatography, followed by direct analysis of the products using gamma-spectroscopy. These results, in conjunction with theory, will be evaluated to generate conclusions relevant to nuclear debris in the shortest time possible.

Log: 183. **IN SITU CHARACTERIZATION OF LI TRANSPORT IN LISN ANODES WITH NEUTRONS.** Liu, D.X. (1); Wang, J. (2); Cao, L. (2); Co, A.C. (1). (1) Department of Chemistry and Biochemistry, The Ohio State University. (2) Nuclear Engineering Program, The Ohio State University.

High energy storage capacity from both anodes and cathodes are required for larger-scale applications. Among other group IV elements, Sn, with its relatively high theoretical specific capacity of 959 mAh/g is a promising alternative anode material. During the alloying/de-alloying process with Li, Sn experiences volumetric expansions of ~300%. This huge volumetric change results in capacity fade. Improvements in the complex architectures such as nanowires, 3D architecture, inactive core/active shell structures are often attributed to enhanced accommodation of the transformations that occur during cycling. However, the transport properties of lithium and their resulting distribution within the material are largely inferred using ex situ and often invasive techniques. A real-time quantification of Li transport using a non-destructive neutron method to measure the Li distribution upon charge and discharge of a Li-ion cell is reported here. In situ Neutron Depth Profiling (NDP) provides temporal and spatial measurement of Li and visualization of its transposition to guide materials development for efficient storage. Using in situ NDP, we probed the onset of lithiation in a model Sn electrode and visualized the enrichment of Li atoms on the surface while monitoring the propagation of Li into the bulk.

Log: 184. **REDUCTION IN BACKGROUND RADIOGENIC SIGNALS IN BULK SEDIMENTS USING SPLITT TECHNOLOGY TO ENRICH  $^{32}\text{Si}$  FOR GEOCHRONOLOGY OF MARINE SEDIMENTS.** Strivens J.(1); Gill G.(2); Brandenberger J.(3); Seiner B.(4); Keillor M.(5); Hossbach T.(6); Finch Z.(7); Arrigo L.(8); and Gillies K(9). (1) PACIFIC Northwest National Laboratory. (2) US Department of Energy.

Isolation and detection of silicon-32 ( $^{32}\text{Si}$ ) ( $t_{1/2}=171.8\pm1.8\text{yrs.}$ ) and its daughter phosphorous-32 ( $^{32}\text{P}$ ) ( $t_{1/2}=14.29\text{days}$ ), will enable the pair to serve as geochronology radioisotopes covering the age-dating gap between lead-210 ( $^{210}\text{Pb}<120\text{yrs.}$ ) and carbon-14 ( $^{14}\text{C}>1000\text{yrs.}$ ). The utility of this measurement is to extend the age modeling of aquatic sediments that integrate the response of the ecosystem to human and climatic changes. This technique requires the enrichment of biogenic silica (BSi) from bulk sediment prior to radiometric detection using an ultra-low-background ( $\sim 10$  counts per day) gas proportional  $\beta$ -detector. By isolating and enriching the BSi fraction of bulk sediments prior to dissolution, it is possible to significantly reduce the background radiation while maximizing the  $^{32}\text{Si}$  content, thereby enabling low background  $\beta$ -detection. In this work, a BSi extraction was performed from bulk sediment, utilizing Stoke's Law via Split Flow Thin Cell Fractionation (SPLITT), post removal of organics and dissolution of carbonates. SPLITT is an expeditive, continuous fractionation method able to divide sediment into two portions; one containing particles above, and the other containing particles below a specific sedimentation coefficient. The sediment fractions from the SPLITT were tested for BSi enrichment via inductively coupled plasma optical emission spectrometry (ICP-OES). Aliquots from each SPLITT step were analyzed for radiogenic nuclide contributions using on an ultra-low-background 14 crystal germanium array. The separation efficiency of the background nuclides was evaluated by comparing the isotope activity in each SPLITT isolation step to the activity of the bulk sediment ( $\sim 18\text{Bq/g}$ ). Background nuclides were also compared to activity in samples prepared using a conventional Sodium Polytungstate separation.

Log: 185. **A system of INAA Automatic Measurement.** Yonggang Yao; Bangfa Ni; Caijin Xiao; Xinghua Wang; Pingsheng Wang; Xiangchun Jin; Long Hua. China Institute of Atomic Energy.

A new INAA (instrument neutron activation analysis) automatic measurement system has been established at China advance research reactor (CARR). This system can be allowed for three sets of detector measurement simultaneously at 24 hours a day. The sample rabbit contains an electromagnetic label which is used for identifying and/or tracing the sample route or location. The automation system is mainly composed of four parts, sample counting system, sample driving system, samples holder and sample changer. Sample counting system contains three gamma spectrometers with counting shielding chamber and electromagnetic label reader, respectively. The sample driving system consisted compressed air and electromagnetic valve. Sample holder is a square board with 12x12 rabbit holes for sample holding. The position of sample to be sent out or received is fixed by moving the board in (x, y) directions. Sample changer is a main part of automation system, which is in charge of receiving sample and allocating the sample to the place which should be. This is controlled by software kingview6.55, programmable logic controller (PLC) and communication module, photoelectric sensor, step motor and its driver.

Log: 186. **DTRA Basic Research for Combating Weapons of Mass Destruction.** Quillen, B.G.; Defense Threat Reduction Agency.

The Defense Threat Reduction Agency safeguards the United States and its allies from weapons of mass destruction (WMD) by providing capabilities to reduce, eliminate, and counter the threat and mitigate its effects. DTRA accomplishes its mission by investing in basic research efforts at universities, national labs and DoD service labs to better counter threats posed by WMD. We also facilitate productive relationships with other scientific organizations and seek to identify promising research efforts overseas. Through the Basic Research program, DTRA recruits and trains scientists and engineers to develop a talented workforce for the future. Basic research is directed towards greater knowledge or understanding of phenomena without regard to specific applications. The DTRA Basic Research Program pursues efforts within and across disciplines such as chemical science, computer/network science, materials science, mathematics, nuclear science, and physics. The presentation will describe the DTRA Basic Research Broad Agency Announcements, research "Thrust Areas," proposal solicitation processes, and technical areas or subjects of interest.

Log: 187. **ANTHROPOGENIC IMPACT ON SGD-ASSOCIATED NUTRIENT FLUXES FROM JAVA, INDONESIA.** Kathrin Hassler (1); Nils Moosdorf (1). (1) Leibniz Center for Tropical Marine Ecology (ZMT).

In recent years submarine groundwater discharge (SGD) received increasing attention as a source of land-derived natural as well as anthropogenic nutrients and other dissolved species to coastal waters. Locally it can exceed riverine fluxes and cause eutrophication of coastal ecosystems. However, SGD in general and anthropogenic impact on SGD-associated nutrient fluxes in particular are still poorly quantified. Java, Indonesia, has a wet climate, regions covered by karstified limestone as well as permeable volcanics and sediments, and is subject to rapid population increase as well as agricultural and industrial development. Hence, on Java, we expect abundant SGD heavily loaded with anthropogenic nutrients and contaminants. The presented project will assess anthropogenic impacts by various methods. We plan to locate groundwater recharge areas with O and H isotopes and possibly REE and Y patterns. Combined with tritium-helium dating of groundwater these may allow for calculating groundwater flow velocities. Source identification (and quantification) of anthropogenic nutrients and other pollutants shall be achieved using multiple isotopic tracers, such as C, N, O, H, S and heavy metal isotopes (Pb, Hg...). We aim at developing a methodology to discern between different anthropogenic contaminant sources (e.g. inorganic fertilizers, septic waste, and animal manure) which is transferable to other locations.

Log: 188. **MERIDIONAL DISTRIBUTION OF FUKUSHIMA-DERIVED RADIOCESIUM IN SURFACE SEAWATERS ALONG A TRANS-PACIFIC LINE FROM THE ARCTIC SEA TO SOUTHERN OCEAN IN SUMMER 2012.** Kumamoto, Y. (1); Aoyama, M. (2); Hamajima Y. (3); Murata A. (1). (1) Japan Agency for Marine-Earth Science and Technology. (2) Fukushima University. (3) Kanazawa University.

Evaluating total amount of radiocesium (Cs-137 or Cs-134) released from the Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident in March 2011 is essential to assess its impact on the marine environment in the North Pacific Ocean. In summer 2012, about one and half years after the accident, we measured the radiocesium activity concentration in surface waters collected in the Arctic Sea, Bering Sea, western North and South Pacific Oceans, and Southern Ocean. The Fukushima-derived Cs-134 was found in the Bering Sea and western North Pacific Ocean between 25 and 63 N degree, which corresponds well to latitudes of main atmospheric deposition of Cs-134 in model simulations. The highest activity concentration, 2.3 mBq/m<sup>3</sup> (not decay-corrected) was observed in the subarctic region (48 N/162 E degree) and not in the transition region between 35 and 40 N degree, in which the FNPP1 is situated (37 N/141 E degree). A synoptic view of Cs-134 concentration activity in the North Pacific revealed that a main body (< 4 mBq/m<sup>3</sup>) of the direct discharged Cs-134 from the FNPP1 was transported westwardly to the international date line along the North Pacific Current around 40 - 45 N degree about one and half years after the accident. A zonal speed of the main body was calculated to be about 12 cm/s. Activity concentration of Cs-137 derived from the nuclear weapon testing and effluents from nuclear fuel processing plants ranged from 0.02 to 1.3 mBq/m<sup>3</sup> along the meridional line.

Log: 189. **DELAYED NEUTRON COUNTING AT CIAE.** Caijin Xiao,Xiangchun Jin,Yonggang Yao,Long Hua, Xinghua Wang,Pingsheng Wang,Bangfa Ni(1).(1) China Institute of Atomic Energy.

Delayed neutron counting (DNC) is a method of uranium determination applied in many fields, such as geology, biology, materials, and nuclear safeguards. Facility of Delayed neutron counting had built at China Institute of Atomic Energy (CIAE) since 1980s. Usually the routine of the method is as following: a sample is irradiated by reactor neutrons for 60 seconds, then transferred to a position waiting for delayed neutron counting, which starts 30 seconds after the end of irradiation for 60 seconds. The routine of DNC may lead to some problems. For example, when the Uranium concentration of a sample is very high some delayed neutron counts would be lost. On the contrary, when the Uranium concentration of a sample is too low, the counting statistics would be very poor. Normalized decaying curve of delayed neutron is proposed to solve the problems. The experiment at Mature Neutron Source Reactor (MNSR) shows that the accuracy of the method is better than 3%.

Log: 190. **USE OF NON DETRUCTIVE ANALYSIS TECHNIQUES FOR THE STRUCTURE DETERMINATION OF URANIUM ORE CONCENTRATES.** Kaye, P., Puxley, C., Poile C. AWE Aldermaston, UK.

Uranium Ore Concentrate (UOC) materials are an easily divertible component of the early stages of the nuclear fuel cycle. As such, they are of great interest to the nuclear forensics community, and much progress has been made in the development of chemical and isotopic signatures for use in determining the provenance of the materials. Given that there are a limited number of refining processes used for the production of UOCs, the ability to determine the uranium compounds has the potential to constrain the number of sources that may be considered when determining material provenance. This presentation will discuss the use of complementary rapid, non-destructive structural analysis techniques, such as powder X-Ray Diffraction, Infra-Red and Raman spectrometry for the characterisation of the uranium compounds in UOC materials to provide supporting evidence for provenance determination.

Log: 191. **Integrated Fukushima Ocean Radionuclide Monitoring (InFORM) Network; Biota measurements.** Mercier J.-F.(1), Chen J (1),Cooke. M.W. (1), Zhang W., Trudel M. (2), Cullen J. (3) (1) Health Canada, Radiation Protection Bureau, Ottawa, Ont Canada (2) Fisheries and Oceans Canada, Pacific Biological Station, Nanaimo, BC Canada (3) School of Earth and Ocean Sciences, University of Victoria, Victoria, BC Canada.

The Integrated Fukushima Ocean Radionuclide Monitoring (InFORM) project is a network involving academic, governmental, and non-governmental organizations, as well as citizen scientists. InFORM is now acquiring data to conduct a thorough radiological impact assessment for Canada's west coast stemming from the Fukushima Daiichi nuclear power plant (FD-NPP) accident, and to effectively communicate these results to the public. Physical models of ocean circulation and mixing predict that predominant currents in the North Pacific will transport a plume of seawater contaminated with FD-NPP radionuclides (dominated by Cs-137 and Cs-134) to Canadian coastal waters in the coming 3-4 years. Models and open ocean measurements agree that the incoming plume should result in a small but measurable increase in the concentration of FD-NPP derived radionuclides. To quantify this increase, and subsequently assess potential environmental and human health impacts associated with it, the InFORM network will collect seawater samples monthly over the next three years from 14 coastal locations between Victoria and Haida Gwaii in British Columbia (BC) and 3 times per year in the North Pacific and Arctic Oceans. Furthermore, approximately 100 biota samples will be collected from BC's coastal waters each year. This presentation will discuss the biota measurements of the first year of this collaboration, collected in the Summer and Fall of 2014.

Log: 192. **APPLICATION OF THERMOGRAVIMETRIC ANALYSIS FOR A HIGH TEMPERATURE COMBUSTION METHOD OF TRITIUM ANALYSIS.** Kim, C.J.; Lim, J.M.; Kang, M.J.; Yang, H.C.; Choi, G.S. Korea Atomic Energy Research Institute (KAERI).

Various laboratories have used a number of different methods to analyze total tritium and OBT. These include oxidizer combustion, oxygen bomb combustion, plasma combustion and a high temperature combustion using tube furnace. In all methods, attention must be paid to two key points: complete combustion and prevention of contamination by ambient atmospheric moisture. One of those, a high temperature combustion using tube furnace is more efficient method for the limited samples and has been used a lot recently. However, there are problem of explosion and incomplete combustion for some environmental samples which include a large amount of organic matter. It cause flowing backward of tritium trapping solution and color quenching. So, this study applied thermogravimetric analysis (TGA) to a high temperature combustion using tube furnace for effective combustion without explosion and incomplete combustion. Ignition points of samples were measured by making use of TGA for some samples which are nuclear waste (activated carbon, oil, resin, etc.) and environmental samples (potato, wheat, sediment, etc.). And, temperature steps of combustion method were determined in consideration of measured ignition points. Finally, we analyzed radioactivity of total tritium and OBT for each samples by combustion method applying the temperature steps. The reported mean values of tritium radioactivity were used to verify this method, and the results were in agreement with assigned radioactivity concentration in reference range. This study shows TGA of some nuclear waste and environmental samples and is aimed to demonstrate the process of stable and rapid combustion method.



Log: 193. **INVESTIGATION OF DEUTERON INDUCED REACTIONS ON nat-Gd UP TO 30 MEV: POSSIBILITY OF PRODUCTION OF MEDICALLY RELEVANT 155-Tb AND 160-Tb RADIOISOTOPES.** Szelecsényi, F.(1); Kovács, Z.(1); Nagatsu, K.(2); Zhang, M.-R.(2); Suzuki K.(2). (1) ATOMKI, Debrecen, Hungary. (2) NIRS, Chiba, Japan.

Excitation function for the nat-Gd(d,x)155-Tb (half-life: 5.32 d) and nat-Gd(d,x)161-Tb (half-life: 6.89 d) nuclear reactions were measured using a stacked-foil activation technique combined with HPGe gamma-ray spectrometry in the deuteron energy range of 30 MeV down to 4.5 MeV. 155-Tb is promising SPECT radioisotope while 161-Tb (b- emitter) can be used for cancer therapy. The measured cross-section data were compared not only with the earlier reported experimental values but also with the results of a theoretical model as well (calculated by the computer code TALYS). The present data show acceptable agreement with the literature values as well as with the calculated excitations functions. Integral yields for the above radio-nuclides and for the contaminating radioisotopes were deduced to evaluate the production circumstances. The results revealed that both 155-Tb and 161-Tb can not be produced in no-carrier-added form even using 100% enriched 155-Gd and 160-Gd targets, respectively. In the case of 155-Tb, the co-produced 156-Tb (half-life: 5.35 d) represents the major contamination up to 14 MeV. The estimated 156-Tb contamination level is around 5 % at 14 MeV. Above this energy the formation of 154-Tb (half life: 22.7 h) also adds to the Terbium contamination level. Since the number of the co-produced and longer-lived 160-Tb nuclides (half-life: 72.3 d) is much higher (around twice) than the 161-Tb in the final product, this production route seems to be useful only for developmental studies. The 160-Tb/161-Tb activity ratio at EOB is around 0.22 using short irradiation time and 14 MeV deuteron beam.

Log: 194. **IDENTIFICATION OF URANIUM-ORE CONCENTRATES FROM MICROMETRIC PARTICLES FOR NUCLEAR SAFEGUARDS AND NUCLEAR FORENSICS BY RAMAN SPECTROMETRY.** Pointurier, F. (1); Ho Mer Lin, D. (2); Marie, O. (1); Manara, D. (2); Moulin, C. (1); Mayer, K. (2). (1) CEA, DAM, DIF. (2) JRC/ITU.

In the framework of nuclear safeguards and forensics, it is of great interest to identify the chemical compositions of uranium ore concentrates (UOC), in order to verify or determine its origin. In some cases, only low amounts of UOC are available, or the sample can be composed of a mixture of several UOCs. Therefore, there is a need to carry out the identification at the individual particle's level. Micro-Raman spectrometry has the ability to carry out structural identification of small-size objects. To evaluate the capacity of this technique for the univocal identification of the different UOC species from minute amounts of sample, particles with sizes ranging from a few micrometers to a few tens of micrometers sampled from five UOCs of known chemical compositions (U<sub>3</sub>O<sub>8</sub>, ammonium diuranate, uranyl hydroxide, sodium diuranate, and uranyl peroxide), production process and origins were analyzed. Results were evaluated with respect to: i) reproducibility of the spectra from one particle to the other for each compound, ii) capacity to distinguish each UOC one from the other, iii) agreement with the results obtained by analyzing the corresponding bulk materials, and iv) agreement with the (scarce) available bibliographic data. Raman bands obtained for both the uranium oxide molecules and the anionic impurities (nitrates, sulfates, etc.) originating from the uranium-ore purification process were considered in this study.

Log: 195. **PLUTONIUM MEASUREMENTS AT ULTRA-TRACE LEVEL IN RECENTLY-DEPOSITED SEDIMENTS IN THE COASTAL RIVERS OF THE NORTH-WEST OF FUKUSHIMA.** Pointurier, F. (1); Evrard, O. (2); Onda, Y. (3); Chartin, C. (4); Hubert, A. (1); Lepage, H. (2); Pottin, A.C. (1); Lefèvre, I. (2); Bonté, P. (2); Laceby, J.P. (2); Ayrault, S. (2). (1) CEA, DAM, DIF, France. (2) CEA/CNRS/UVSQ, LSCE/IPSL, France. (3) University of Tsukuba, Japan. (4) Université catholique de Louvain, Belgium.

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident led to important releases of radionuclides into the environment and trace levels of plutonium (Pu) were detected in northeastern Japan. However, measurement of Pu isotopic atom and activity ratios is required to differentiate between the contributions of global nuclear test fallout and FDNPP emissions. In this study, we measured Pu isotopic ratios in recently deposited sediments along rivers draining the most contaminated part of the inland radioactive plume. For this, we carried out a thorough chemical purification and concentration of Pu from sediment samples (5 g dry material) and precise isotopic measurements using a double-focusing sector field ICP-MS. Results showed that the entire range of measured Pu isotopes (i.e.,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$ ) were detected in all samples, although in extremely low concentrations. The  $^{241}\text{Pu}/^{239}\text{Pu}$  atom ratios measured in sediment deposits (0.0017–0.0884) were significantly higher than the corresponding values attributed to the global fallout ( $0.00113 \pm 0.00008$  on average for the Northern Hemisphere between  $31^\circ$ – $71^\circ\text{N}$ ). The results indicated the presence of Pu from FDNPP, in slight excess compared to the Pu background from global fallout, representing up to ca. 60% of Pu in the analyzed samples. These results demonstrate that this radionuclide has been transported relatively long distances (45 km) from FDNPP and deposited in rivers representing a potential source of Pu to the ocean.

Log: 196. **Occurrence of Organically Bond Tritium and  $^{14}\text{C}$  in the Mohelno Lake system.** Svetlik, I. (1); Fejgl, M. (2); Povinec, P.P. (3); Simek, P. (1). (1) Nuclear Physics Institute AS CR. (2) National Radiation Protection Institute. (3) Department of Nuclear Physics and Biophysics, Faculty of Mathematics, Physics and Informatics, Comenius University.

Two Nuclear Power Plants (NPPs) Temelín (ETE) and Dukovany (EDU) are operated in the Czech Republic. Light water pressurized reactors (LWPR) with the installed power output 2x1000 MW and 4x440 MW, respectively, are employed in both of them.  $^{14}\text{C}$  (in gaseous effluents) and  $^3\text{H}$  (HTO in liquid releases) are responsible for dominant effective doses from all radionuclides discharged NPPs during the standard operation. Likewise, a minor part of produced  $^{14}\text{C}$  is released by liquid waste and enters into the connected water systems. Liquid discharges from the EDU NPP are released by outlet drainage canal into the creek “Skryjský creek”, which flows into the Mohelno reservoir about 50-60 meters far from the water inlet of the NPP. The short distance between these two places causes a “loop” effect. Deep valley of the Mohelno Lake reduces possibility of ventilation. Experiments aimed on OBT (organically bond tritium, non-exchangeable forms) occurrence in the plants around the Mohelno Lake were performed in 2011. OBT activity levels enhanced by one magnitude above background values were proved in the Mohelno Lake vicinity. A more detailed sampling campaign was realized in 2014 with the aim on the study of  $^{14}\text{C}$  and  $^3\text{H}$  distribution at this interesting system of the Mohelno Lake.

Log: 197. **RAPID ISOLATION OF NEPTUNIUM FROM SOLUTION AND SOIL.** Rosenberg, B. (1); Shozugawa, K. (2); Steinhauser, G. (1). (1) Colorado State University. (2) The University of Tokyo.

Actinides are great sentinels for ascertaining the condition of nuclear fuel elements following the meltdown of a nuclear reactor. Isotopic and activity ratios of environmental plutonium significantly different from the background ratios established by nuclear explosions indicate that there was a breach in the fuel elements of a reactor. Measuring these ratios employ mass spectrometry and alpha spectrometry. We propose using Np-239 as a sentinel; the gamma rays emitted from short-lived Np-239 have intensities and energies that make detection of this radionuclide rapid by gamma spectroscopy. Potential problems with spectroscopic analysis are interferences by volatile radionuclides, such as radiotellurium and radioiodine. Therefore, the goal was to isolate Np-239 by ion specific extraction chromatography using Eichrom® resin columns. UTEVA, RE, TRU, and Actinide resins were evaluated for their ability to isolate volatile radionuclides from neptunium. UTEVA and RE resins loaded and eluted with nitric acid of varying concentrations can isolate neptunium from more than 99% of the iodine in a sample and effectively exclude tellurium and cesium. This exclusion was observed in both aqueous and soil matrices. Therefore, following an accident, rapid evaluation of reactor core conditions can be assessed by isolating Np-239 using extraction chromatography on soil or rainwater samples and detecting this radionuclide with gamma spectroscopy.

Log: 198. **Amidoxime Group Modified Wool Fiber as a High-efficient Adsorbent for Recovery of Uranium from Seawater.** Bai-Hua Chen; Chong Cheng; Ning Wang; Jie Xiong; Jun Wen; Tao Jiang; Sheng Hu. Institute of Nuclear physics and chemistry, China Academy of Engineering Physics, Mianyang, China.

Adequate uranium resources are very important for the sustainable future development of nuclear power. In the past years, uranium extraction from seawater has attracted extensive research interests. The amidoxime-based synthetic fibrous adsorbents are considered to be the most suitable adsorbents. A lot of natural wool products are disposed of as waste every year. Wool fibers have been used as economical adsorbents to uptake heavy metal ions from industrial waste water. In this paper, natural wool, instead of synthetic fiber, functionalized with amidoxime (Wool-g-AO) has been used as a new adsorbent for uranium (VI) adsorption from seawater. The amidoxime (AO) groups based wool fibers were prepared by a co-radiation-induced graft copolymerization of acrylonitrile and acrylic acid and the subsequent conversion of cyano groups to amidoxime groups on the graft chains. Some experimental parameters such as surface pre-treatment and Ray energy, total dose, solution ratio were varied to optimize the polymerization. The physicochemical properties of Wool-g-AO were investigated using infrared spectroscopy (IR), scanning electron microscope (SEM) and X-ray photoelectron spectroscopy (XPS). Results showed that the ligand monomers were successfully grafted onto the wool fibers. The adsorbent showed above 300 % graft rate of the ligand monomers. The adsorption experiments were carried out in both artificial seawater and the natural ocean environment. In contrast with the synthetic polymer adsorbents, wool fibers, composed of  $\alpha$ -keratin, have much greater hydrophilicity, which imparts it more chance to contact with uranium ions in solutions. The results showed the adsorption capacity of Wool-g-AO is 62.62 mg/g in artificial seawater and 0.3 mg/g in the natural ocean environment respectively, much better than the Amidoxime-Polyethylene polyethylene fiber sorbent (47.54 mg/g in artificial seawater and 0.15 mg/g in natural seawater, respectively).

Log: 199. **SEPARATION OF AN(III) AND LN(III) IN NITRIC ACID MEDIUM USING ISOHEX-BTP/SIO<sub>2</sub>-P ADSORBENT.** Yuezhou Wei; Shunyan Ning; Xinpeng Wang; Ruiqin Liu. Shanghai Jiao Tong University.

2,6-bis(5,6-diisohexyl)-1,2,4-triazin-3-yl)pyridine) (isoHexyl-BTP) is a nitrogen-donor chelating ligand which shows high extraction selectivity for An(III) over Ln(III) in nitric acid medium. We synthesized a porous silica/polymer-based isoHexyl-BTP adsorbent (isoHexyl-BTP/SiO<sub>2</sub>-P) for the separation of An(III) from Ln(III) in HLLW. The adsorption of isoHexyl-BTP/SiO<sub>2</sub>-P towards <sup>241</sup>Am(III), <sup>239</sup>Pu(IV) and middle-heavy Ln(III) (Am, Eu, Gd, Dy) in 0.01-4 M HNO<sub>3</sub> increased as the nitric acid concentration increased. The uptake rate of <sup>241</sup>Am(III) was  $\geq 99\%$  when nitric acid  $\geq 2$  M. The adsorption ability towards Ln(III) increased as atomic number increased until Ho(III) and SFAm/Eu reached 88 in 2 M HNO<sub>3</sub>. isoHexyl-BTP/SiO<sub>2</sub>-P showed relatively poor adsorption towards <sup>241</sup>Am(III) and almost no adsorption towards Ln(III) in the solutions of 0.99-3.99 M NaNO<sub>3</sub> with pH 2. To explain the adsorption differences between nitric acid and nitrate media and the different adsorption ability towards Ln(III) and An(III), the work focused on the

isoHexyl-BTP/SiO<sub>2</sub>-P adsorption mechanism in nitrate medium and coordination chemistry of Ln(III) with the adsorbent by using extended X-ray absorption spectroscopy (EXAFS), acid base titration and ion chromatography characterization methods. It was found both H<sup>+</sup> and NO<sub>3</sub><sup>-</sup> directly participate in the coordination reaction during the adsorption process. For the middle-heavy Ln(III), M(isoHexyl-BTP/SiO<sub>2</sub>-P)<sub>3</sub>(NO<sub>3</sub>)<sub>3</sub>•nHNO<sub>3</sub> was supposed as the main adsorbed species. The Ln(III)-N(isoHexyl-BTP/SiO<sub>2</sub>-P) bond length of the first coordination layer had the decrease tendency as the atomic number increased, which can explain to some extent the increase adsorption affinity of isoHexyl-BTP/SiO<sub>2</sub>-P towards Ln(III) in concentrated nitric acid as the atomic number increased.

Log: 200. **CURRENT TRENDS OF CS-137 CONCENTRATIONS IN COASTAL WATERS NEAR THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT.** Hirose, K. Sophia University.

As a result of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in 2011, large amounts of radionuclides have been released in marine environment. Marine environmental radioactivity monitoring by TEPCO and NRC has been continued since 2011. In 2014, concentrations of FDNPP-derived Cs-137 in open ocean surface waters off Fukushima generally decreased to less than 0.005 Bq/L. On the other hand, sporadic higher Cs-137 concentrations in seawater, being more than 1 Bq/L, still occurred near north and south outlets of the FDNPP. In order to control environmental discharge of radionuclides, it is important to elucidate causes of sporadic increases of Cs-137 in coastal seawater near the FDNPP. We, here, examined the relationship between Cs-137 peaks and rainfall events during the period of March to September 2014 (AMeDAS data provided by Japan Meteorological Agency). The result revealed that the peaks of the Cs-137 concentrations in coastal waters near the FDNPP appeared to follow heavy rainfall events. Possible processes to produce the Cs-137 peaks in coastal waters due to rainfall events are direct input due to atmospheric deposition of Cs-137 and indirect processes such as surface runoff of rainwater sweeping contaminated area, increasing discharge from trenches as a result of heavy rain and increasing discharge via groundwater.

Log: 201. **SEPARATION AND ANALYSIS OF URANIUM IN THE SRM IAEA-384 AND IAEA-385.** Han, J. (1); Hu, S. (1); Yang, C. T. (1). (1) Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics.

The IAEA reference material can be used for assurance/quality control of the analysis of radionuclides in the environment, as well as for the development and validation of analytical methods. Furthermore, the isotope ratio may provide a valuable contribution to environmental monitoring. Herein, the pretreatment, separation, and analysis of the IAEA -384 and 385 samples have been done. Quantitative analysis values of uranium measured by TIMS were  $1.37\text{E-}4$  (234U), 0.0161 (235U), 2.26 (238U) ug/g in IAEA-384, and  $1.24\text{E-}4$  (234U), 0.0164 (235U), 2.30 (238U) ug/g in IAEA-385. The massic activities of uranium isotopes were  $42.1408 \pm 0.19$  (234U), 1.7284 (235U),  $37.5892 \pm 0.19$  (238U) Bq/Kg in IAEA-384, and  $27.1015 \pm 0.05$  (234U), 1.3166 (235U),  $29.2706 \pm 0.06$  (238U) Bq/Kg in IAEA-385, respectively. The results were in line with the certified mass activity of CRM (dw 95% confidence interval). The isotope ratios of 235U and 238U in the environmental soil samples were in good agreement with natural Uranium.

Log: 202. **SPATIAL AND TEMPORAL CHANGES OF Cs-137 CONCENTRATIONS DERIVED FROM NUCLEAR POWER PLANT ACCIDENT IN RIVER WATERS IN EASTERN FUKUSHIMA, JAPAN DURING 2012–2014.** Ochiai, S. (1); Ueda, S. (1); Hasegawa, H. (1); Kakiuchi, H. (1); Akata, N. (1,2); Ohtsuka, Y. (1); Hisamatsu, S. (1). (1) Institute for Environmental Sciences. (2) National Institute for Fusion Science.

The spatial and temporal changes of Cs-137 concentrations in river water samples collected from seven rivers in areas contaminated highly by the Fukushima Dai-ichi Nuclear Power Plant accident were investigated to reveal the dominant factors controlling concentrations under base-flow conditions. River water samples were collected once a year at 16 points in the seven rivers under base-flow conditions from 2012 to 2014 in eastern Fukushima Prefecture, Japan. After each sample was filtrated, the Cs-137 concentration in dissolved and particulate forms was measured by gamma-ray spectrometry. Mean Cs-137 concentrations were 0.16 (range: 0.01–0.59) Bq/L and 0.19 (0.01–0.60) Bq/L in dissolved and particulate forms respectively in 2012, and concentrations decreased to 0.069 (0.01–0.27) Bq/L and 0.099 (0.002–0.53) Bq/L in 2014, respectively. Previous studies have reported that concentrations in a dissolved form are dependent on the mean Cs-137 inventories in each watershed of each sampling point as evaluated from airborne surveys. Our three year observation revealed that the slope of the regression line for dissolved concentrations on the inventories in each sampling year continuously decreased from 2012 to 2014. This result shows that the mobility of dissolved Cs-137 from watersheds to river water decreased with time. In addition, in 2014 some sampling points exhibited a faster decline of dissolved Cs-137 concentrations than other sampling points, which implies that the mobility of dissolved Cs-137 decreased faster in these watersheds between 2013 and 2014. This work was performed under contract with the government of Aomori Prefecture, Japan.

Log: 203. **A SENSITIVE AND SELECTIVE ELECTROCHEMICAL SENSOR FOR THE DETERMINATION OF URANYL IONS BASED ON RGO/CALIX[6]ARENE MODIFIED ELECTRODE.** Cheng, C.M.; Wang N.; Jiang, T.; Hu, S. Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics.

In the present work, a new type of mercury-free, sensitive and selective electrochemical sensor based on reduced graphene oxide (rGO)-calix[6]arene (SC6) modified glassy carbon electrode was described for the detection of uranyl ions. Graphene oxide (GO) nanoplates were functionalized with water-soluble 4-sulfonated calix[6]arene (SC6) via a self-assembly strategy, and highly water-dispersed GO-SC6 nanocomposites were prepared. The resulting GO-SC6 nanocomposites were used to fabricate GO-SC6 modified glassy carbon electrodes (GO-SC6-GCE) by using a simple drop casting method. And then, the GO on the electrode was electrochemically reduced at a potential range of 0.0 to -1.5 V (vs. SCE) to obtain rGO-SC6 modified glassy carbon electrode (rGO-SC6-GCE). Due to the selective affinity of SC6 towards uranyl ions as well as the outstanding conductivity of rGO, rGO-SC6-GCE displayed excellent electrochemical response to uranyl ions. The fabrication of modified electrodes is simple and highly reproducible, demonstrating its promising application in routine uranyl ions analysis.

Log: 204. **Fluorescent BINOL-Based Sensor for Thorium Analysis.** Jun Wen (1), Sheng Hu (1), Xiao-Lin Wang (1). (1) Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics.

Because of the widespread use of thorium and its toxic properties, the development and improvement in analysis methods for the determination of thorium would be useful. We developed a novel 1,1'-bi-2-naphthol (BINOL) derivative fluorescence sensor L-1 for the recognition of thorium ion with a fluorescence quench response. This ligand showed high selectivity and sensitivity for thorium ion recognition. When an equivalent of Th<sup>4+</sup> was added to the solution of L-1, dramatic fluorescence quenching (quenching efficiency: 64%) was observed, suggesting that compound L-1 showed a specific response with Th<sup>4+</sup> ions due to the chelation-enhanced fluorescence quenching (CHEQ) effect. This is the first one-to-one stoichiometric responding chemical sensor for thorium, and indicated a 1:1 bonding mode between L-1 and Th<sup>4+</sup> ions (Figure 2). The detection limit [4] of L-1 for the determination of Th<sup>4+</sup> was estimated to be  $6 \times 10^{-7}$  M in 1:1 MeOH:H<sub>2</sub>O (v/v). Moreover, the binding constant (K) derived from the fluorescence titration data was found to be  $3.4 \times 10^3$  using a Benesi-Hildebrand plot.



Log: 205. **The evaluation of Submarine Groundwater Discharge (SGD) input toward the Yatsushiro Inland Sea by combining the  $^{222}\text{Rn}$  spatial distribution and temporal fluctuations.** Nikpeyman, Y. (1); Hosono, T. (2); Ono, M. (3); Yang, H. (1); Shimada, J. (1); Takikawa, K. (4). (1) Department of Earth Sciences, GSST, Kumamoto University. (2) Priority Organization for Innovation and Excellence, Kumamoto University. (3) The National Institute of Advanced Industrial Science and Technology, Japan. (4) Center for Marine Environment Studies, Kumamoto University.

Estimation of the total submarine groundwater discharge (SGD) as a path by which solutes and nutrients travel from terrestrial areas to coastal areas.  $^{222}\text{Rn}$ , as a powerful geochemical tracer, can detect SGD signals in coastal seas. Although, many researchers have used this tool, detailed discussion of the estimation of multiple source contributions is still rare in interpretations of observed  $^{222}\text{Rn}$  data. This study aimed to make a connection between spatial  $^{222}\text{Rn}$  distribution in the study area (towing results) and the temporal  $^{222}\text{Rn}$  fluctuations (mooring results) observed in the Yatsushiro Sea, southwestern Japan that is the largest inland sea in Japan. Several rivers with high  $^{222}\text{Rn}$  concentrations discharge into the study area that cause SGD flux overestimation. Initially, the seawater spatial  $^{222}\text{Rn}$  distribution pattern evaluated using the multi-detector towing. In addition, river grab samples analyzed for  $^{222}\text{Rn}$  concentrations. Then, the Yatsushiro Sea model was used to evaluate the  $^{222}\text{Rn}$  distribution originated from 15 flow in rivers and the results subtracted from the towing output to determine the SGD patches.  $^{222}\text{Rn}$  moorings were operated simultaneously with seawater salinity monitoring at a representative point for each SGD patch during summer neap-tide and winter spring-tide periods. Fundamentally, rivers discharging close to the mooring sites were the least important functions on observed  $^{222}\text{Rn}$  concentration for all points, although they cause seawater salinity perturbations. Finally, by multiplying the SGD advection rates and the SGD patch areas, the total SGD volume calculated for the whole Yatsushiro Sea (2.73 MCM/day). The total SGD is evaluated to be 1/6 to Yatsushiro Inland Sea to the average river discharge.

Log: 206. **RADIOACTIVITY MONITORING OF ENVIRONMENTAL SAMPLES AROUND A RESEARCH REACTOR : EFFECTS OF FUKUSHIMA ACCIDENT.** Kang, M. J.; Kim, W. Y.; Kim, C. J.; Park, D. W.; Lee, C. W. (Korea Atomic Energy Research Institute).

A research nuclear reactor has been operated at KAERI in Korea for neutron beam application and irradiation tests of the materials and nuclear fuels. Radioactivity monitoring of environmental samples around the research reactor has been performed to confirm the nuclear safety of surrounding environments by the operation of nuclear facilities. The environmental radioactivities on gross  $\alpha$ , gross  $\beta$ , uranium radioisotope, H-3, Sr-90, and  $\gamma$ -radionuclides were analyzed routinely in various samples such as air particulates, an iodine air filter, air moisture, soil, sediment, pine needle, rainwater, surface water, underground water, fallout, farm products, and livestock products. The radioactivity of  $\gamma$ -radionuclides was measured using a gamma spectrometer (Ortec, HPGe detector with MCA). The  $\gamma$ -radionuclides includes the artificial nuclides such as Cs-134, Cs-137, I-131, Co-60, Cr-51, Fe-59, Mn-54 and natural ones such as Be-7 and K-40. In the iodine air filter sampled during 1 week at March or April of 2011, I-131 radioactivity was measured with a range of 0.124~1.78 mBq/m<sup>3</sup>. This was caused by an Fukushima accident in Japan. In the air particulates collected during March or April, I-131 radioactivity was measured with a range of 0.108~0.158 mBq/m<sup>3</sup>. Cs-134 and Cs-137 radioactivity was also measured with range of 0.0582~0.124 and 0.0639~0.117 mBq/m<sup>3</sup>, respectively. In rainwater sampled during April of 2011, I-131, Cs-134, and Cs-137 radioactivity was measured as very low concentration. After June, the I-131, Cs-134, and Cs-137 radioactivity became below MDA in iodine air filter, air particulate, rain water, and fallout.

Log: 207. **Quantification of submarine groundwater discharge and associated nutrient input with radium and radon isotopes on the tropical island of Java (Indonesia).** Oehler, T. (1); Moosdorf, N. (1). (1) Leibniz-Zentrum für Marine Tropenökologie.

Submarine groundwater discharge (SGD) occurs widespread in many coastal regions, but global estimates about the impact of SGD on coastal nutrient, carbon and freshwater budgets are scarce. A junior research group investigating SGD from tropical islands on a global scale started recently at the Leibniz-Centre for Tropical Marine Ecology (ZMT, Bremen). The group will investigate the tropical island of Java (Indonesia) in order to improve our understanding of processes governing SGD from tropical islands. The island of Java is one of the most densely populated areas worldwide struggling with groundwater pollution from high nutrient loadings. Due to permeable rocks and high annual precipitation rates, high rates of freshwater discharge and the associated nutrient fluxes are expected. Based on satellite infrared images and a hydrogeological model of Java, several sampling sites will be selected at which SGD will be quantified with radium and radon isotopes. The sampling sites will represent different coastal regions featuring different types of SGD. In the south-eastern part of Java a focused flow along fractures can be expected in a karst area. In the northern part, SGD might be forced by tidal pumping in sediments of coastal plains. The dominant mechanisms, forcing submarine freshwater discharge (e.g. hydrological gradients, geological setting) will be identified and used for extrapolation. The amount of nutrients entering the coastal regions of Java by SGD will be quantified and used for mass budgets. Here we present the project plan in order to seed cooperation and exchange experiences.

Log: 208. **OPTIMIZATION OF COMPTON SUPPRESSION FOR AIR FILTER ANALYSIS.** YOHO, M.D; LANDSBERGER, S.; University of Texas.

Compton suppression systems for low level radioactivity counting either for neutron activation analysis, environmental background or fission product identification is now a well-established methodology existing in many word-wide laboratories. The metric to establish the efficiency of the system is the peak-Compton (P/C) ratio of the 662 keV photopeak of Cs-137. The P/C ratio has been reported to be anywhere from 500-1100 depending on the electronics, germanium detector resolution, size of (NaI) or BGO shield, etc. While the detector is in the shield there is an optimum placement of the source to achieve the best P/C ratio which is usually calculated on the detector and then every 0.5-1.0 cm away from the detector up to 10 cm or more away. We have shown that by placing the Cs-137 source every 0.25 cm one can calculate a significantly better P/C ratio which then can be used specifically for air filters which have inherently very thin height dimensions. A evaluation of the system and its application for air filters will be presented.

Log: 209. **PROMPT GAMMA RAY ANALYSIS OF CRUDE OIL SAMPLES.** Khiari F. Z (1); Isab A H.(2); Khateeb-ur-Rehman(1).(1)Department of Physics ; (2) Department of Chemistry, King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia.

The quality of crude oil is evaluated through its carbon/hydrogen (C/H) concentration ratio. The lower this ratio is, the higher is the quality of the crude oil. Prompt gamma ray analysis was carried out to determine C/H concentrations of crude oil samples. The measurements were carried out using a 14 MeV neutron based PGNA setup using a LaBr<sub>3</sub>:Ce detector. Prompt gamma ray spectra were recorded from several crude oil samples and standards. Carbon concentrations of the samples were determined through the measured intensity of 4.44 MeV gamma rays while hydrogen concentration was determined through the measured intensity of 2.22 MeV gamma rays. Water and benzene samples were used to calibrate the PGNA setup. The carbon to the hydrogen (C/H) elemental ratio, measured from the crude oil samples in the present study, varies from 1.403 to 1.847. It is in agreement with results of (C/H) ratios of the samples obtained through chemical analysis.

Log: 210. **FAST FLUX LOSS IN SOIL SAMPLES DUE TO THEIR HYDROGEN CONTENTS- AN ESTIMATION.** Naqvi A. A (1); Khateeb-ur-Rehman(1); Isab A. H. (2). (1) Department of Physics ; (2) Department of Chemistry, King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia.

Hydrogen contents in soil samples affect the prompt gamma analysis of hydrocarbon in soil samples. Such studies are important for environmental studies. Monte Carlo simulation were carried out to calculate yields of hydrogen, carbon, oxygen, and silicon prompt gamma rays from soil. The simulated PGNA setup mainly consists of a cylindrical sample placed at a distance of 10 cm (center-to-center) distance from the 14 MeV neutron generator target. A cylindrical gamma ray detector detects prompt gamma rays emitted at 90 degrees with respect to the neutron beam axis. Gamma ray yields were calculated for 1.78, 4.44, and 6.11 MeV prompt gamma rays of silicon, carbon and oxygen, respectively. Thermal neutron capture gamma ray yields were calculated for 3.55 MeV gamma rays of silicon and 2.22 MeV gamma rays of hydrogen. All gamma rays yields were calculated for different moisture concentrations in the soil samples. It has been found that an increase in hydrogen concentration in the sample results in (a) an increase of the intensities of 2.22 MeV gamma rays of hydrogen and 3.55 MeV gamma rays of silicon due to an increase in thermal neutron flux, and (b) a corresponding decrease in intensities of 1.78 and 6.11 MeV prompt gamma rays due to loss in fast neutron flux. The increasing trend of thermal capture gamma rays or decreasing trends of NIS gamma rays can be used to estimate the fast flux losses due to hydrogen concentration of soil samples in NIS induced prompt gamma ray analysis of soil samples.

Log: 211. **SPECTROELECTROCHEMICAL SENSOR FOR DETECTING TECHNETIUM IN STORED NUCLEAR WASTE AND THE ENVIRONMENT.** Heineman, W.R. (1); Branch, S. (1); Crawford, A. (2); Bryan, S.A. (2). (1) University of Cincinnati. (2) Pacific Northwest National Laboratory.

A need exists for chemical sensors to monitor technetium compounds in stored nuclear waste and in the surrounding environment at the Hanford Site in Washington. A novel sensor that combines electrochemistry, spectroscopy, and selective partitioning into a film has been developed to meet the stringent demands of these applications. The sensor consists of an optically transparent electrode (OTE) coated with a selective film. Sensing is based on the change in optical signal (absorption or fluorescence) for attenuated total reflectance at the OTE which accompanies electrochemical modulation of analyte that has partitioned into the film. Selectivity for the analyte relative to other sample components is obtained by choice of film material, electrolysis potential, and wavelength for optical monitoring. Spectroelectrochemical sensors have adequate selectivity and ruggedness for applications that involve complex samples in harsh environments such as the determination of technetium compounds in nuclear waste stored at the Hanford Site. The sensors are also suitable for environmental applications such as monitoring pertechnetate leaking from storage tanks into subsurface water at the Hanford Site.

Log: 212. **VARIABILITY ANALYSIS OF NATURAL DECAY CHAIN AND NEUTRON INDUCED BACKGROUND FOR QUALITY ASSURANCE IN HPGE SPECTROSCOPY.** Yoho, M.D.(1);Porterfield, D. (2), Landsberger, S. (1). (1) University of Texas, (2) Los Alamos National Laboratory.

Twenty-one HPGe laboratory background spectra were collected from October 2012 to June 2014 at Los Alamos National Lab. Intensities from the U-238 and Th-232 natural decay series both above and below the Radon gas progeny were analyzed for temporal variability. It was found that the U-238 decay products, including those in secular equilibrium with Ra-226, demonstrated minimal temporal variability beyond what would be expected from counting statistics. The Th-232 daughters displayed variability at most twice what would be expected from counting statistics. Further, variability of thermal and fast neutron flux was found to be minimal from an analysis of the 139 keV Ge-74 (n, gamma) and Ge-72 (n, n') peaks. Further, no statistically significant dependence between elevated background levels and season was found. Finally, background spectral line intensities were compared to values previously reported in the literature.

Log: 213. **Radiochemical Neutron Activation Analysis In A Sample Containing Ta, W, Au, And Pt In The Presence Of Fission Products.** Morrison, S.S. (1,2); Clark, S.B. (1), Eggemeyer T.A. (2), Finn, E.C. (2), Hines, C.C. (3), King, M.D. (3), Metz, L.A. (2), Morley S.M. (2), Seiner, B.N. (2), Wall D. E. (3). (1) Department of Chemistry, Washington State University. (2) Pacific Northwest National Laboratory. (3) Nuclear Radiation Center, Washington State University.

Activation of gold (Au) has been used to estimate neutron fluence resulting from criticality events, but analysis is complicated by simultaneous production of other gamma-ray emitting fission products. Confidence in neutron fluence calculations can be increased by quantifying additional activation products such as tantalum (Ta), tungsten (W) and platinum (Pt), in addition to Au. This work describes a radiochemical separation procedure designed for the quantification of Ta, W, Au, and Pt activation products for a sample in which fission products are also present. The separation procedure employed a series of anion exchange chromatography columns to separate anionic complexes of Ta, W, Au and Pt from fission products. This procedure was evaluated with samples composed of nominally  $10^{12}$  fissions, 4  $\mu\text{g}$  of 93% enriched uranium and  $10^9$  atoms of Ta-182, W-187, Pt-197, and Au-198. Analysis results following the separation procedure developed in this work demonstrated chemical yields exceeding 90%. The minimum detectable activities were improved by 2-3 orders of magnitude for all activation product radionuclides.

Log: 214. **A Sensor Composed of CdTe Nanoparticles for Radiation Detection.** Cutright, J. (1); Wei, H.T. (2); Huang, J.S.(2); Cao, R.L. (1). (1) The Ohio State University. (2) University of Nebraska-Lincoln.

Highly sensitive radiation detection is an effective way of locating special nuclear materials. Recent advances in ultraviolet photodetectors show that nanocomposite layers comprised of ZnO nanoparticles and the organic semiconducting polymer P3HT can be used to outperform traditional inorganic semiconductor devices. The outstanding performance of the new photodetector is the result of interfacial charge trapping of electrons in the type-II semiconductor heterojunction between the nanoparticles and the P3HT. In dark the device functions as a photodiode with a Schottky rectifying contact. Under irradiation charge trapping of the electrons in the device allows for a large hole current to pass, effectively making it a photoconductor with an Ohmic contact. A device structure composed of nano composite CdTe is fabricated and evaluated in terms of its electronic properties and response to x- and gamma-rays. Rather than using ZnO nanoparticles, CdTe nanoparticles are dispersed throughout the P3HT. CdTe's high cross section for  $\gamma$  interaction makes it a good choice for the electron-hole pair donor. This work focuses on characterizing the efficiency of the novel CdTe x-ray and gamma radiation detector, simulating the photon interactions with the device in MCNP, and measuring the stopping power of the device.

Log: 215. **A SOLID PHASE EXTRACTION MICROFLUIDIC SYSTEM FOR ACTINIDE ANALYTICAL TRACING ELEMENTAL ANALYSIS.** Jun Gao, Benjamin T. Manard, Alonso Castro, Dennis P. Montoya, Ning Xu and Becky Chamberlin Los Alamos National Laboratory, Los Alamos, NM.

Microfluidic technology is a rapidly growing multidisciplinary field that provides great potential in separation, detection and reaction applications, including advanced radiochemical separation and actinide detection. The highly sophisticated microfluidic systems can be integrated using multiple functional micro-chips, also coined "Lab-on-the-chip". The actinide separation can be performed on the chip, and actinide and trace impurities can be harvested to enable highly sensitive quantification. The conventional ion exchange column-based actinide separations are commonly performed on milliliter scale. Here, we utilize the state-of-art microfluidic technology to develop micro-columns (20-100 $\mu$ L) for actinide separation. These independent and novel microscale column modules fabricated by laser cutting can be further integrated into more sophisticated platform for complicated sample preparation and actinide separations, thus, enabling the high throughput. These microfluidic technologies offer such advantages as: 1) availability of a cost-effective platform with robust sampling statistics, which can readily and inexpensively screen a large number of actinide samples in a rapid fashion, 2) ultra-low consumption of actinide samples, hazard reagents, and reduced radioactive waste production from analyses, 3) the micro columns between 20 to 100  $\mu$ L, can provide effective reaction networks to retain, elute and separate actinide analytes, 4) cost-effectiveness, and 5) miniaturized footprint designs are near-infinitely flexible and reproducible. In our pilot study, the separation efficiencies of plutonium, uranium, and trace elements were evaluated by micro-columns separation and ICP-OES detection. The results suggested that our ion exchange microfluidic separation system could reduce not only the actinide sample consumption to as low as  $\sim$ 200  $\mu$ L for the tracing elemental determination, but also the amount of radioactive waste generation. (LA-UR-14-29072)

Log: 216. **BASIC EXAMINATION OF A MOLYBDENUM-99/TECHNETIUM-99M PRODUCTION SYSTEM USING AN ELECTRON LINEAR ACCELERATOR.** Sekimoto, S.(1); Tadokoro, T.(2); Kani, Y.(2); Ohtsuki, T.(1). (1) Kyoto University Research Reactor Institute. (2) Hitachi Ltd.

Most of molybdenum-99 (Mo-99), which is the generator of Technetium-99m (Tc-99m) for such as single photon emission computed tomography (SPECT), has been produced through U-235 (n,f) reaction by using research reactors in several countries. Some of those research reactors have operated continuously more than 40 years and then, they shall be closed down due to their deterioration in the near future. Shortage of Mo-99 resulting from the shutdown or no operation of research reactors can be a global issue. Recently, Mo-99-production by using an accelerator has been investigated in Japan. This investigation has been promoted since domestic production of Mo-99 is required in Japan without depending on importation of Mo-99 as in the past. To insure a stable supply of Mo-99 for hospitals, Mo-99-production system in several or more facilities in Japan is also expected. In this study, we commenced to examine the new complex where Mo-99-production system by using an electron linear accelerator and Tc-99m-separation system by sublimation method are furnished. Our goal is to provide the complex that is as compact and inexpensive as possible. In our experiments, molybdenum-100 oxide powder was irradiated with bremsstrahlung photons. The irradiation was carried out using the electron linear accelerator at Kyoto University Research Reactor Institute. We will present the optimization of the electron energy and current for Mo-99-production. The ability of the sublimation method for Tc-99m-separation will be discussed.

Log: 217. **HIGH SENSITIVE ANALYSIS OF RADIOACTIVE CESIUM IN JAPANESE FOODSTUFFS: THREE YEARS AFTER THE FUKUSHIMA ACCIDENT.** Shozugawa, K.(1); Saito, T.(2); Hori, M.(1); Matsuo, M.(1). (1) University of Tokyo. (2) Akita radiation measuring station(Beguredenega).

In April 2012, to achieve further food safety and consumer confidence, Japanese government revised the regulatory limits of radioactive materials in general foodstuffs to reduce maximum permissible dose from 5 mSv/year (500 Bq/kg) to 1 mSv/year (100 Bq/kg). We analyzed Cs-134, Cs-137 and K-40 in 50 kinds of foodstuffs in supermarket including processed foods with high sensibility over three years after Fukushima accident. Milk, polished rice, yoghurt, Japanese tea, salmon, cereal, blueberry, miso, and apple had a trace Cs-134 and Cs-137 from  $10E-3$  to  $10E-0$  Bq/kg, however, some mushrooms which were bought outer Fukushima prefecture included radioactive cesium over regulatory. In view of the detection of Cs-134 which has a short half-life of 2.06 year and the Cs-134/Cs-137 ratio, we can conclude that Cs-137 in food detected in remote area 250 km or more from Fukushima nuclear plant originated in Pre-Fukushima such as Chernobyl accident (1986) and atmospheric nuclear explosions (from 1945).



Log: 218. **THE PROGRESS OF CARR INAA FACILITIES OF CHINA.** Bangfa Ni, Caijin Xiao, Yonggang Yao, xinghua Wang, Long Hua, Xiangchun Jin, Pingsheng Wang, Guiying Zhang(1).(1) China Institute of Atomic Energy.

Three kinds of NAA facilities have been installed at China Advance Research Reactor (CARR) with 60MW of maximum power and  $8 \times 10^{14}$  n.cm<sup>-2</sup>.s<sup>-1</sup> neutron flux has been established, which include conventional NAA, thermal neutron PGAA and cold neutron PGAA, respectively. Two irradiation channels are allocated for short irradiation and long irradiation of conventional NAA with the neutron flux of  $3 \times 10^{14}$  n.cm<sup>-2</sup>.s<sup>-1</sup> and the medium temperature about 40°C at sample positions. Pneumatic systems are used for sample transfer. The speed of rabbits is designed to be 20 m/s, and it takes 3 s to travel from reactor to detector. It can be used for determination of very short lived nuclides, such as <sup>18</sup>F. A new INAA automatic counting system has been established. The characteristics of the system include 1) the sample rabbit contains an electromagnetic label which is used for identifying and/or tracing the sample route or location. 2) Three sets of detector mounted the electromagnetic label reader can be measuring simultaneously. 3) All the steps of automation measurement are monitored by computer and any abnormality information will be sent to user's mobile. The Thermal neutron PGAA channel is designed in tangent direction from the heavy water reflector of CARR and filtered by a diameter of 60x200mm Bismuth single crystal. The neutron flux is expected to be about  $1 \times 10^8$  n.cm<sup>-2</sup>.s<sup>-1</sup> at sample position with a neutron chopper mounted. The cold neutron PGAA source is cooled by liquid hydrogen. The flux is expected to be about  $1 \times 10^7$  n.cm<sup>-2</sup>.s<sup>-1</sup> at sample position. The delayed neutron counting equipment and neutron depth profile facility will be also accompanied.

Log: 219. **SGD SNIFFER - AN AUTONOMOUS GAMMA-SPECTROMETER MONITORS SUBMARINE GROUNDWATER DISCHARGE TRENDS IN HAWAII.** Dulaiova, H. (1); Kamenik, J. (2); Waters, C. A. (1); Kennedy, J. (1). (1) University of Hawaii Department of Geology and Geophysics. (2) Nuclear Physics Institute of the Academy of Sciences of the Czech Republic.

On the Kona coast of the Hawaii Island almost 100 percent of the terrestrial runoff is in form of submarine groundwater discharge. Yet, little is known about the variability of these groundwater fluxes on longer time scales and their response to long-term sea level rise and climate change. To address this gap, we developed the SGD Sniffer, the first autonomous gamma-spectrometer deployed for long-term coastal SGD monitoring. The instrument represents a significant improvement over previous submarine gamma-spectrometers in that it is very robust, has high-sensitivity, and is completely autonomous. We will present the technical parameters of the new instrument as well as data collected over its 9-month deployment. It has collected seasonally variable data and was operational through several processes that affect SGD, yet we have never had direct observations on their effect: a tsunami (Apr 16, 2014), a tropical cyclone (Iselle, August 8, 2014) as well as significant westerly wind anomalies over the central and eastern Pacific which resulted in unfavorable precipitation conditions in the fall of 2014.

Log: 220. **ULTRA-TRACE ICP-MS ANALYSIS OF PLUTONIUM AND RADIOCESIUM ISOTOPES FOR TRACING THE FUKUSHIMA NUCLEAR ACCIDENT RELEASED RADIONUCLIDES IN THE ENVIRONMENT.** Zheng, J. (1); Tagami, K. (1); Bu, W.T. (2); Uchida, S. (1). (1) National Institute of Radiological Sciences. (2) Peking University.

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident led to the release of large amounts of radionuclides into the environment. This accident has renewed considerable public concern about the dangers posed by radioactive contamination in the environment and the related radiation exposure from the contaminating radionuclides. Sensitive and accurate analysis of radiation sources and understanding the amount and the range of radioactive contamination are essential for effective radiation protection. Since the FDNPP accident, gamma ray emission fission products, such as Cs-134, Cs-137 and I-131 have been analysed using gamma spectrometry in environmental and food samples to estimate the contamination range and to prevent internal radiation exposure resulting from ingestion of contaminated food. Mass spectrometric techniques, especially inductively coupled plasma mass spectrometry (ICP-MS) have played important role in the studies of source identification of radioactive contamination and long-term environmental behaviour of released radioactive materials released from the FDNPP accident. Here, we report the new development of SF-ICP-MS and the novel ICP-MS/MS for accurate determination of radioactive Pu isotopes and radiocesium isotopes in environmental samples collected in Fukushima Prefecture after the accident. We identified the isotopic compositions of radioactive Cs and Pu isotopes released from the FDNPP accident, and estimated the released amounts of Cs-135 and Pu isotopes. We demonstrated that radiocesium in the terrestrial environment was mainly released from the Unit 2 reactor. The potential applications of the obtained FDNPP-sourced Cs and Pu fingerprints in the study of long-term environmental behaviour of the released radioactive materials will also be discussed.

Log: 221. **ADVANCES IN THE PGAA-ACTINIDE PROJECT.** Kudejova, P.(1); Revay, Z.(1); Genreith, C.(2); Rossbach, M.(2); Randrimalala, T.(2). (1) MLZ, Technische Universität München. (2) IEK-6, Jülich Research Center.

At the last MARC IX conference, we have reported a start-up project for validation of thermal neutron capture cross-section data of actinides, namely  $^{237}\text{Np}$ ,  $^{242}\text{Pu}$  and  $^{241}\text{Am}$ . Now - three years later - the results and comparison to the results in some evaluation databases (e.g. ENDF, JENDL or JEFF) will be presented together with the description of the performed experiments at the high-intensity cold neutron PGAA instrument at the research reactor FRM II of the Heinz Maier-Leibnitz Zentrum (MLZ). In frame of the PGAA-Actinides project a new fast neutron PGAA instrument (spectrum mean by ca. 2 MeV) was put into operation at FRM II with the aim of measuring prompt gamma spectra following e.g. (n, n') or (n, p) reactions of investigated actinides. First test of the instrument and the instruments itself will be shortly introduced.

Log: 222. **CONVENTIONAL AND EPITHERMAL INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS OF EDIBLE JAPANESE SEAWEEDS FOR MULTIELEMENT CONTENT.** Fukushima, M. (1); Isaac-Olive, K. (2); the late Nakano, Y. (3); Chatt, A (2). (1) Department of Food and Environment, Faculty of Science and Engineering, Ishinomaki Senshu University, Minamisakai, Ishinomaki, Miyagi 986-8580, Japan. (2) Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, 6274 Coburg Road, Room 212, P.O. Box 15000, Halifax, NS, B3H 4R2, Canada. (3) Research Reactor Institute, Kyoto University, Sennann, Osaka, 590-0494, Japan.

Edible Japanese seaweeds are known to contain a number of elements. There is an increasing interest to estimate the average daily dietary intake of these elements. Fresh Japanese tangle and sea mustards were collected from the ocean, washed with tap water, cut into several pieces, freeze dried, and pulverized. Dry fir needles, glue plant and hijiki were purchased from a local market and pulverized. One portion of the seaweed samples was irradiated at the Dalhousie University SLOWPOKE-2 reactor (DUSR) facility for the determination of Br, Ca, Cl, I, K, Mg, Mn, Se, Sr, and V by epithermal instrumental neutron activation analysis in conjunction with anticoincidence counting (EINAA-AC). Another portion was irradiated at the Kyoto University reactor (KUR) for the determination of As, Co, Cr, Fe, Na, Rb, Sb, Sc, and Zn by conventional INAA. Reference materials were analyzed to validate the methods. Both total and bioaccessible fraction of the elements were estimated. Details of the method and result will be presented.

Log: 223. **Integrated application of radioisotope and aerial infrared imagery for detection of SGD from a volcanic island.** Eunhee Lee(1); Kil-Yong Lee(1); Heesung Yoon(1); Dong-Chan Koh(1); Kyoochul Ha(1). (1) Groundwater Department, Korea Institute of Geoscience and Mineral Resources.

Submarine groundwater discharge (SGD) has been recognized as a significant pathway of groundwater and chemical species to the coastal zone. In this study, aerial thermal imagery combined with radioisotope tracers were used to reveal the pathway of SGD to coastal water body in the southern shore of Jeju Island, South Korea. Sea surface temperature (SST) map of coastal water was obtained from UAV (Unmanned Aerial Vehicle) thermal data to detect the location of SGD. Geochemical data including  $^{222}\text{Rn}$  activity were collected to assess the effect of groundwater discharge on nearshore water body. SST map successfully captured the cold groundwater plumes and the temperature signals by groundwater discharge were reduced as groundwater disperses into the coastal ocean. Rn activities of coastal water showed strong correlation with SST. These comparisons revealed the pathway of discharged groundwater to the coastal water body can be tracked, which provide valuable information for mass balance of Rn activity and the estimation of SGD.

Log: 224. **DEVELOPMENT OF AN IRIIDIUM DISSOLUTION METHOD FOR THE EVALUATION OF POTENTIAL RADIOLOGICAL DEVICE MATERIALS.** Hodgson, A.P.J.(1,2); Grimes, R.W.(2); Jarvis, K(3); Marsden, O.J.(1). (1) AWE plc, Aldermaston, Reading, Berkshire, RG7 4PR, United Kingdom. (2) Department of Materials, Imperial College London, Royal School of Mines, Exhibition Road, London SW7 2AZ, United Kingdom. (3) Centre for Environmental Policy, Faculty of Natural Sciences, Imperial College London, South Kensington Campus, London, SW7 1NA, United Kingdom.

iridium-192 sources are generated in nuclear reactors through the irradiation of stable target materials. However, variation in: neutron energy, flux and irradiation time; target material characteristics and purity; the activation cross sections of the desired reactions; decay and daughter progeny in-growth; and any post irradiation processing, can play a key part in determining the isotopic and chemical composition of the material produced. These isotopic ratios, together with those of any activated elemental impurities, therefore have the potential to provide information relating towards not only the material's production date, but also the source's production route, irradiation history and original elemental and isotopic composition. A computational study to evaluate the effect that neutron flux spectra have on radionuclide production within such materials has indicated a number of potential signatures. These signatures show significant variability depending on the reactor conditions employed, but need to be validated using materials of known production history prior to further utilisation. As part of this process, a new method is therefore being developed to enable the dissolution of iridium sources without biasing the signature of the embedded impurities. Details of the technique being used will be provided, and the various issues that have been encountered during development and how these will impact on future source evaluations will be discussed. © British Crown Owned Copyright 2014 /AWE

Log: 225. **STUDY OF DIFFERENT METHODS USING THE PORTABLE LIQUIDE SCINTILLATION COUNTER TRIATHLER FOR ON-SITE MEASUREMENTS.**

Landstetter, C.(1); Zapletal, M.(1); Katzlberger C.(1). (1) Austrian Agency for Health and Food Safety, Department of Radiation Protection and Radiochemistry, Spargelfeldstrasse 191, 1220 Vienna, Austria.

The potential occurrence of small scale incidents has been discussed in Austria during the last 10 to 20 years. A document about on-site measurements and sampling plans following a small scale incident has been created in 2011. Currently just in-situ gamma spectrometry measurements, the local dose rate and measurements using a contamination monitor are performed. In case of the presence of low energy gamma emitters or pure alpha and beta emitters wipe tests have to be sent to the laboratory for measurements. This reduces the speed of reporting the analytical result. Therefore on-site estimations or determinations of the involved radionuclides and their activity concentration are preferred. 2013 a project was funded by the Federal Ministry of Agriculture, Forestry, Environment and Water Management to evaluate the usability of the portable liquid scintillation counter Triathler TM for such purposes. During the first phase of this project the Triathler TM was used in the laboratory and gross alpha and beta measurements and wipe tests were performed. These measurements were compared using accredited methods usually used in the laboratory. The separation of the alpha and beta emitters via pulse length index using different scintillation cocktails was tested. Background measurements with different scintillation cocktails using different counting modes were performed and the occurrence of luminescence after sample preparation in the lower energy regions was evaluated. In this paper the results of the project are summarised.

Log: 226. **FURTHER INVESTIGATION OF EPITHERMAL NEUTRON ACTIVATION ANALYSIS IN THE DETERMINATION OF HEAVY METALS IN THE ARCTIC ATMOSPHERE.** Lara, R.; Landsberger, S.; University of Texas.

In the past years our laboratory has successfully employed epithermal neutron activation analysis (NAA) with and without Compton suppression to determine a range of heavy metals including arsenic, antimony, indium, etc. As part of an Environment Canada 34 year study to evaluate heavy metals in the winter months at Alert in the Canadian Arctic, we have further explored the use of these methodologies to establish the presence barium, potassium, silicon, silver, strontium and zinc. Silver was determined using cyclic epithermal NAA with a 60 second irradiation, a 10 second decay and a 30 second counting time using Ag-110 and its 25 second half-life. After a 10 minute irradiation and a 5 minute decay and a 10 minute counting time silicon was determined using Si-29 and its 6.5 minute half-life. After 5-10 air filters were counted they were placed in an automated Compton suppression sample changer to count the remaining radionuclides with a 6-12 hour decay and a 1.5 hour counting time. Potassium was determined using K-42 and its 12.8 hour half-life; barium was determined using Ba-139 and its 1.4 hour half-life, strontium was determined using Sr-87m and its 2.8 hour half-life; and zinc was determined using Zn-69m and its 12.8 hour half-life. All these NAA activation products employed Compton suppression with the exception of Ag-110. An evaluation of typical concentrations, uncertainties and typical detection limits will be given.

Log: 227. **QUALITY CONTROL IN THE ANALYSIS OF RADIOACTIVITY FROM THE OIL EXPLORATION SECTOR.** Landsberger, S.(1); Graham, G.(2); (1) University of Texas (2) Enviroklean Product Development Inc.

The presence of radioactivity in the production of oil is now a well-known phenomenon. Specifically Ra-226, Rn-222, Ra-228 and Pb-210 and their various decay products are of environmental concern especially to the workers in the field. The Nuclear Engineering Teaching Lab at the University of Texas has been in collaboration with Enviroklean Product Development Inc. an environmental restoration and chemical product company in clean-up of technologically enhance naturally occurring radioactive material (TENORM) in the oil sector. In particular radioactive scale build-up in the equipment has been an on-going concern. Unlike typical NORM samples, Ra-226 and its daughters dominate the radioactivity and as such is not in any secular equilibrium with any of the radionuclides above the decay chain that emanate from U-238. An effort has been undertaken to establish quality control procedures and to produce an internal reference material. Scale build-up has a significant amount of iron, barium and strontium and as such self-attenuation of photons, especially the 46.5 keV gamma-ray from Pb-210, can drastically underestimate the radioactivity concentration. A procedure was developed to prepare an internal house-standard using only a 20-g sample of scale obtained from west Texas. An evaluation of the homogeneity, counting procedures and determination of the self-attenuation factors will be discussed.

Log: 228. **SIMULATIONS OF BACKGROUND CHARACTERISTICS OF HPGe DETECTORS OPERATING UNDERGROUND USING THE MONTE CARLO METHOD.**

Breier, R. (1); Povinec, P.P. (1); Hamajima, Y. (2). (1) Department of Nuclear Physics and Biophysics, Comenius University in Bratislava; (2) Low-level Radioactivity Laboratory, University of Kanazawa.

An increasing number of experiments have been devoted to the detection of very rare events, e.g. in neutrinoless double beta-decay searches, in dark matter and neutrino experiments, as well as in environmental physics. Their common feature is a utilization of high sensitive low-level counting spectrometers, operating very often deep underground. If a detector was constructed from selected radioactivity free materials, the dominating background component in a surface or shallow underground laboratory would be cosmic rays. The flux of soft component cosmic-ray particles (electrons, positrons, gamma-rays) can be considerably decreased due to electromagnetic showers in materials with high atomic number, e.g. lead, iron, copper, etc. On the other hand, the flux of hard component particles (muons) can be decreased only by installation of detectors deep underground, or by using an anti-cosmic (anticoincidence) shielding. For the background optimization of a counting system it is useful to use a Monte Carlo simulation, so the background characteristics can be estimated before constructing a counting system. The aim of the present work has been a developing of a computing code that would allow compute background components of low-level HPGe  $\gamma$ -spectrometers optionally equipped with anti-cosmic shields. The simulation is based on the CERN's GEANT 4 code, and it will be carried out for the 290 m w.e. Ogoya underground laboratory in Japan and for the 4800 m w.e. laboratory in Modane.

Log: 229. **Rapid and Simultaneous Detection of Alpha/Beta Radioactivity in Food by Solid Phase Extraction - Liquid Scintillation Counting.** Lin, Zhichao; Healey, Stephanie; Wu, Zhongyu. Winchester Engineering and Analytical Center, U.S. Food and Drug Administration.

A simple and rapid radioanalytical method for screening alpha and beta radioactivity in a wide variety of foods was developed and validated. DGA resin (N,N,N',N'-tetra-n-octyldiglycolamide, Normal) with high affinity for actinides and rare earths including americium (Am), plutonium (Pu), and yttrium (Y), was used to extract these radionuclides from food matrices in a mixture of 8 M HNO<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> resulting from conventional wet acid digestion. The retained radionuclides were stripped from the resin using 0.1M HCl – 0.1M H<sub>2</sub>C<sub>2</sub>O<sub>4</sub>, converted into chlorides after oxidizing the oxalate, and then mixed with Ultima Gold AB for discriminative quantification of gross alpha/beta activities by liquid scintillation counting. Dairy, meat, vegetable, grain, and complex meal samples ranging from 30 to 100 grams and spiked with varying alpha/beta activities were tested to develop a robust extraction procedure and optimize instrument settings. The study showed that 95±5 % of actual sample activity was recovered in 10-min batch extraction using 1 g of DGA resin, and negligible inter-sample quench variability allowed >90% counting efficiency without developing chemical/color quench curves. The method was found to be capable of detecting gross alpha and beta activity at ~0.06 Bq/kg and ~0.4 Bq/kg, respectively. The measured results were typically within ±15% of known values with a reproducibility of ~20% for various foods spiked with ~1 Bq alpha/beta activity. The procedure's simplicity allowed for the completion of 8 sample analyses within ~5 hrs and made it suitable for screening a large number and wide variety of foods in the event of a nuclear or radiological emergency.



Log: 230. **GAMMA3 A NEW VERSATILE GAMMA RAY SPECTROMETER DEDICATED TO LOW LEVEL DETECTION OF FISSION PRODUCTS IN ENVIRONMENTAL SAMPLES.** G. Douysset, G. Le Petit, A. Cagniant, P. Gross, J.P. Fontaine, O. Delaune and S.Topin. CEA/ DAM/DIF.

Detection of trace amounts of fission product is of particular importance in various fields of environment monitoring such as treaty verification, nuclear forensics or site survey. A multi-crystal HPGe surface-level spectrometer, named Gamma3, has been designed at our laboratory, it is now fully operational. The setup is composed of three high efficiency/high resolution Canberra BEGe5030P spectrometers implemented in a configuration allowing high geometrical detection efficiency for photons emitted by various types of samples from thin aerosol filters to radioactive gas cell or large volume bottles. The device is fitted with state-of-the-art features to reduce the background: enhanced passive shield (low/very low activity lead, borated polyethylene layer, clean N<sub>2</sub> gas injection) and an integral active muon veto. Long term background signal has been studied extensively; it is shown that the integral count rate is as low as 3.0 counts per minute in the 20-2500 keV range which is among the lowest published background for a surface level instrument. Data acquisition is performed by a dedicated multichannel 14 bits digital system, this configuration produces listmode files that are processed offline using home-developed ROOT-based routines. This feature allows to process signals produced by the detectors in various modes: single, additive, coincidence/anti-coincidence mode. Coincidence (double/triple) mode can be used to detect weak amounts of fission products for which direct gamma signatures might be buried in the Compton background produced by an active sample. For measurement of radioactive gases the system can be fitted either with a double sided standard gas cell or with a home designed system comprising two embedded silicon detector operating in electron/photon coincidence mode. Performances of the Gamma3 system will be illustrated for various types of samples: from compressed aerosols filters or environmental air samples to solid and liquid matrixes.

Log: 231. **COMPARISON OF CHROMATOGRAPHIC SEPARATIONS OF NP FROM BULK PU AND U MATRICES.** Doyle, J. L.(1); Trujillo, J. A.(1); Townsend, L. E.(1); Schake, A. R.(1); Porterfield, D. R.(1); Tandon, L.(1). (1) Los Alamos National Laboratory.

Neptunium (Np) quantification in special nuclear material, specifically plutonium (Pu) and uranium (U) matrices, can provide information on material production (such as reactor conditions, separation conditions, etc.), but typically Np concentrations in these matrices are present at trace or ultra-trace levels. The analytical techniques generally applied to measure total neptunium content include mass spectrometry methods and radiochemistry (alpha and/or gamma spectrometry). For both of these methods the primary matrix (U or Pu) introduces potential spectral interferences, and therefore a novel separation method is required. There are many published examples of methods to separate and measure Np in low level environmental samples containing Pu and U. However, literature on the separation of Np from bulk Pu and U matrices is scant. Current separation procedures used in the actinide analytical chemistry group at Los Alamos National Laboratory can successfully separate Np from bulk plutonium, but the method has roughly a 10 ppm limit of detection. This radiochemical method produces excellent results for Np quantification in Pu, however this procedure is time consuming and utilizes hazardous chemicals and methods. At LANL, for bulk U there is no validated radiochemical method for determination of Np in a U matrix. A novel, state of the art method is being developed to improve method detection limits, yields, and decontamination factors. The new proposed methods utilize novel radiochemical separation schemes (anion exchange and extraction chromatography) for Np from bulk Pu and U the results for which will be presented.

Log: 232. **Creating Benchmark Configurations for Radioisotope Identification Algorithm Challenges.** Myers, S.C.; Lombardi, M.L.; Los Alamos National Laboratory.

Implementing a successful Global Nuclear Detection Architecture (GNDA) requires proficient radiation sensors and infrastructure to detect, identify and discriminate threat from non-threat materials. Currently, most detectors have significant failings in performing real time nuclide identification (ID). The Domestic Nuclear Detection Office of the Department of Homeland Security organized the algorithm improvement project to challenge and improve Rad/Nuc ID algorithms with open and closed benchmark data sets that include a prioritized list of radioactive material configurations. Over 400 high-quality benchmarks have been measured to date with the following gamma ray detection media: high purity germanium, sodium iodide, lanthanum bromide, cadmium-zinc-telluride, and cesium-lithium-yttrium-chloride (CLYC). The benchmark configurations encompass a broad range of common real world conditions in all nuclide categories, including a thorough set of challenges in the special nuclear material/threat category. It is hoped the commercial sector will gain a greater understanding of which real world configurations an ID algorithm needs to handle, and also to provide vendors with a means to readily evaluate revisions and upgrades. Software has been developed that will enable vendors to create their own synthetic spectra for testing and development efforts.

Log: 233. **GAMMA RAY ISOTOPIC ANALYSIS OF HEAT SOURCE PLUTONIUM (PU-238) USING THE FRAM ISOTOPIC CODE.** Myers, S.C. (1); Porterfield, D.R. (1); Carver, N.R. (1); Jump, R.K. (1); Foster, L.A. (1). (1) Los Alamos National Laboratory. Heat source plutonium (>80% Pu-238) is used to make radioisotope thermoelectric generators that provide power and heat on a variety of spacecraft deployed by NASA in the fulfillment of their mission. The requirement to determine the plutonium isotopic abundances in the feedstock materials is currently performed using Thermal Ionization Mass Spectrometry (TIMS) at the Chemistry, Metallurgy and Research (CMR) building at Los Alamos National Laboratory (LANL). There are plans to phase out the TIMS capability at CMR and a need exists for a reliable replacement methodology. An alternative method for the determination of plutonium isotopic abundances uses high-purity germanium (HPGe) detectors to acquire gamma-ray spectra from sealed sample containers. In this study, 1 to 2 gram samples were analyzed using the LANL developed FRAM isotopic routine. There are a number of published reviews evaluating the accuracy and precision of FRAM applied to weapon grade plutonium, but few such studies have been done on heat source plutonium. This report will compare TIMS and FRAM results for 18 heat source plutonium samples using gamma ray spectra acquired with a 25% coaxial HPGe detector. The 18 samples were measured during two separate campaigns designed to demonstrate this capability and optimize the technique for future work. In addition to the calculations of plutonium isotope mass fractions (i.e., Pu-238 through Pu-242), we also explored the feasibility of using the spectral data to determine the concentrations of the following subset of radionuclides that are often present in such samples: Am-241, Np-237, Pu-236, Th-228, Th-232 and U-234.

Log: 234. **Thermodynamics relevant to f-element separations: Old areas and new frontiers.** Braley, J.C. (1) Colorado School of Mines.

If all equilibria relevant to a given separation are accurately accounted for, the effectiveness of a given separation can be predicted by thermodynamic data. The ability to predict, or model, a given separation is imperative for industrial scale separations relevant to used nuclear fuel. This presentation will provide a high-level analysis of thermodynamic information available in the current literature with the goal of identifying 1) significant gaps and 2) areas where further assessment is not urgent. A particular focus will be placed on thermodynamic information available for the heaviest actinides, actinides in less accessible oxidation states and ligands most relevant to f-element separations ("soft" donors, selective adjacent lanthanide reagents, etc). Applications of these reagents in used nuclear fuel cycle separations and nuclear forensics applications will be considered.

Log: 235. **USING KINETIC PHOSPHORESCENCE ANALYSIS TO QUANTIFY EUROPIUM AND TERBIUM.** Beck, Chelsie (1); Seiner, Brienne (2); Smith, Steven (3); Finch, Zachary (4); Friese, Judah (5). Pacific Northwest National Laboratory (1) US Department of Energy (2).

Rapid screening and analysis for lanthanides is important to a variety of fields including; nuclear fuel chemistry, geochemistry, and environmental science. However, the analysis is often challenging due to the similar chemical properties of the lanthanides. These similarities complicate elemental separations and result in interferences using common quantitative analytical techniques. Kinetic Phosphorescence Analysis (KPA) offers several potential advantages for the analysis of the phosphorescent lanthanides (i.e. europium, terbium, dysprosium, thulium and samarium) including non-destructive analysis and improved luminescence measurements by means of a pulsed laser source with time resolution. Using ethylene diamine tetraacetic acid (EDTA) as the complexing agent, only terbium and europium have quantifiable lifetimes. This study focuses on identifying the optimal conditions to measure terbium and europium by KPA; including the ideal matrix and EDTA concentration. The limit of detection and linear response range for each of the elements, as well as the ability to measure europium and terbium in the presence of potential interferents, is discussed.

Log: 236. **ELEMENTAL CONSTITUENTS IN AEROSOLS COLLECTED FROM PLUTONIUM PRODUCTION ENCLOSURES.** Xu, N.; Schappert; M.; Montoya, D.; Martinez, A.; Martinez, P.; Tandon, L. Los Alamos National Laboratory.

Aerosol particle samples were collected from the air, floor, and outbound exhaust ventilation of plutonium production glove boxes with substrates such as PVC filter, cotton swipes and glass based HEPA filter. Various analytical methods for dissolving aerosol particles for trace elemental analysis were evaluated. The element constituents in particulate samples were determined by the ICP-MS and ICP-OES methods. Elemental analysis for plutonium contained particles is not trivial. The three major challenges include 1) complete/near complete dissolublization of the elements including plutonium into aqueous solution; 2) the contamination issue related to the sample preparation process; and 3) the element background in the collector media. We have demonstrated in our study that pressurized closed vessel acid digestion method yielded the highest dissolution efficiency while circumventing elemental contamination, and was employed for particle dissolution from PVC and glass HEPA filters. Open vessel complete acid digestion, on the other hand, appeared to be the most effective in dissolving cotton swipes. LA-UR-14-29107

Log: 237. **DIRECTIONAL ELPASOLITE DETECTOR SYSTEM FOR MEASUREMENTS OF NEUTRON AND PHOTON FLUX.** Barzilov, A. (1); Guckes, A. (1); Guss, P.P. (2). (1) University of Nevada Las Vegas. (2) Remote Sensing Laboratory - Nellis.

Neutron and photon measurement technologies are essential to fulfilling homeland security and nonproliferation mission areas including detection and localization of missing or smuggled radiological or nuclear materials. To address these needs, a radiation detection system based on the scintillation elpasolite compound  $\text{Cs}_2\text{LiYCl}_6\text{:Ce}$  (CLYC) was studied for simultaneous measurements of neutron and photon flux. The system, consisting of an array of CLYC detectors, allowed directional measurements with the localization of radiation sources. Computational modeling of elpasolite scintillator responses shows that the three-detector array is feasible to determine the direction to a neutron source and a gamma-ray source. The prototype detection system consisting of three CLYC detectors equipped with the digital data analysis unit was deployed for directional measurements, confirming the computational results. A detector energy resolution of 5% was measured for 662 keV photons. The on-the-fly pulse shape discrimination was utilized to separate neutron and gamma-ray waveforms with a figure of merit of 2.3 in experiments with a plutonium-beryllium source. The tests were carried out for different combinations of gamma-ray and neutron sources positioned at different coordinates. The measured data were processed through a maximum-likelihood estimation algorithm providing a direction to radioactive sources. The results of the experiments will be discussed. **ACKNOWLEDGMENT** The authors acknowledge the professional staff of RMD, Watertown, Massachusetts, for the production of the detectors, for providing these detectors to the Remote Sensing Laboratory, and for their support and advice. This work was done by National Security Technologies, LLC, under Contract No. DE-AC52-06NA25946 with the U.S. Department of Energy and supported by the Site-Directed Research and Development Program.

Log: 238. **Separation of Lanthanides and Actinides in Supercritical Fluid Carbon Dioxide.** Wai, C.M. (1); Mincher, B.J. (2); Fox, R.V.(2); Quach, D.L.(2); Yen, C (1); Mincher, M.E. (1). (1) University of Idaho. (2) Idaho National Lab.

Using supercritical fluid carbon dioxide (sc-CO<sub>2</sub>) as a solvent for dissolution and extraction of lanthanides and actinides from solid materials is a well-established technique in the literature. This non-traditional extraction technique offers a number of advantages over conventional acid dissolution and organic solvent extraction/separation processes. Minimizing secondary liquid waste generation, effective penetration into solid matrices, rapid separation of solute from solvent, and containment of volatile products in a closed system are some unique features of this novel extraction technique. The technology has many potential applications in metals and energy related industrial processes and associated environmental problems. Some potential applications of the sc-CO<sub>2</sub> technology of current interest include treatment of nuclear wastes and recycle rare earth elements. Separation of lanthanides and actinides in sc-CO<sub>2</sub> after their dissolution/extraction is a key issue for these applications. It is known that TBP-nitric acid complexes such as TBP(HNO<sub>3</sub>)<sub>1.8</sub>(H<sub>2</sub>O)<sub>0.6</sub> are soluble in sc-CO<sub>2</sub> and are capable of dissolving lanthanide oxides (Ln<sub>2</sub>O<sub>3</sub>), UO<sub>2</sub>, and PuO<sub>2</sub> directly in sc-CO<sub>2</sub>. In the CO<sub>2</sub> dissolution process with TBP(HNO<sub>3</sub>)<sub>1.8</sub>(H<sub>2</sub>O)<sub>0.6</sub>, the oxides of all lanthanides and actinides are dissolved in the CO<sub>2</sub> phase as their nitrate-TBP complexes. On-line separation techniques are necessary to recover lanthanides, uranium and other actinides dissolved in the CO<sub>2</sub> stream. This paper describes our current research and development activities in achieving lanthanides and actinides separations after their dissolution in sc-CO<sub>2</sub> using water/sc-CO<sub>2</sub> counter-current stripping techniques. Some results of lanthanides and actinides separations in water/sc-CO<sub>2</sub> counter-current systems based on their differences in distribution coefficients with respect to aqueous acidity, redox chemistry, and ligand chelation capability are presented.

Log: 239. **A COMPARISON OF ELUTION PROFILES FOR SR SEPARATIONS USING COMMERCIALLY AVAILABLE EXTRACTION CHROMATOGRAPHY PRODUCTS.** McLain, D.R.(1); Mertz, C.J.(2); Sudowe, R.(1). (1) University of Nevada, Las Vegas. (2) Argonne National Laboratory.

As a result of terrorist activities over the course of the last two decades, a number of previously unconsidered threats have emerged for federal and state authorities, including the use of radiological dispersal devices. These types of devices are much simpler to build and much more "cost effective" than fabricating a nuclear device because they contain material that is generally much easier to obtain than fissile material. One of the key impact factors of an RDD attack is the quantity and type of radiological material employed. One of the important isotopes when discussing RDDs is the pure beta emitter strontium-90. Quantification of strontium-90 in a post-detonation situation would be an important part of accomplishing several goals, including dating the age of the radiostrontium source and determining the total amount of strontium-90 released in the attack. There are many procedures that have been developed to isolate strontium from environmental samples over the years, and it is likely that a variation of one of these would be used in quantifying

the strontium in the area. The majority of the modern methods developed employ extraction chromatography with commercially available Sr Resin to carry out the actual separation. This study investigates whether there are any differences in the elution profiles or strontium recovery levels of several commercially available extraction chromatography products.

Log: 240. **MATRIX EFFECTS OF DISSOLVED STEEL ON EXTRACTION CHROMATOGRAPHIC STRONTIUM SEPARATIONS.** McLain, D.R.(1); Amato, V.(1); Sudowe, R.(1). (1) University of Nevada, Las Vegas.

Most current radioanalytical protocols have been developed for the analysis of air, water, soil and bioassay samples. While these protocols build the foundation of operational environmental monitoring, they are not necessarily suitable for the analysis of samples that will be encountered after a nuclear incident. Due to their simple and "cost effective" design, it is likely that such an incident would involve a radiological dispersal device. A device of this nature would use an easily obtainable material like strontium-90. In case of such an incident it will be important to determine the amount of strontium-90 present in the affected area to ensure the appropriate response. This objective requires procedures that can be used to rapidly and accurately separate and determine radioactive materials from matrices found in urban or metropolitan environments. Of particular interest are methods that can be applied to the analysis of concrete, steel, and glass. Chromatographic resins have been used in radioanalytical separations for several years now, and are likely to be used in such methods. A large amount of data has been published on these resins' retention capabilities for a variety of elements found in environmental samples. Little can be found, however, on the effects of debris matrices likely to be encountered in a post detonation scenario. If the effects of matrix constituents could be quantified, ways to streamline the separation procedures may arise. To this end, standard reference material steel has been examined to understand how its composition affects the uptake of strontium to Sr Resin.

Log: 241. **ACCELERATOR MASS SPECTROMETRY: RECENT DEVELOPMENTS AND APPLICATIONS AT CENTA.** Povinec, P.P.; Comenius University in Bratislava.

Accelerator Mass Spectrometry (AMS) laboratory has been established at the Centre for Nuclear and Accelerator Technologies (CENTA) at the Comenius University in Bratislava comprising of a MC-SNICS ion source, 3 MV Pelletron tandem accelerator, and an analyzer of accelerated ions. A preparation of targets for  $^{14}\text{C}$ ,  $^{129}\text{I}$  and Pu isotopes AMS measurements is described shortly. The  $^{10}\text{Be}$  analyzing method using a switching magnet at  $45^\circ$  as an analyzing magnet, and a double-anode ionization chamber with silicon nitride stack absorber for  $^{10}\text{B}$  isobar suppression is described. Using this simple method a detection limit for  $^{10}\text{Be}/^{9}\text{Be}$  mass ratio of the order of 10-12 has been achieved. A development of AMS technique for potassium, uranium and thorium analysis of materials required for ultra-low background underground experiments is briefly discussed as well.



Log: 242. **LOW LEVEL GAMMA-RAY COUNTING IN OGOYA UNDERGROUND LABORATORY.** Hamajima, Y. Kanazawa University.

In Ogoya Underground Laboratory (OUL), where the overburden is 270 m w.e., we have 10 well, 6 planar, and 1 coaxial low background Ge detectors for low-level radioactivity measurement. All detectors have large active volume and provide excellent counting efficiency. Ultra low-background aluminum is used for the end-cap. The cryostat is the J-type and/or U-type and the preamplifier is located outside of lead shield. The lead shield is 15 to 25 cm thick, and its upper part is covered by 10 to 15 cm of iron. The inner 3 to 5 cm of the lead shield is made of lead refined 200 years ago or Oxygen-Free Copper. The nitrogen gas from the Dewar is blown to the top of the end cap. Due to this shielding conditions, the background count rates of OUL detectors are about 1/100 of the aboveground ones. I will report some significant low level gamma-ray measurements in OUL, i.e. the detection of low level cosmogenic radio nuclides in a few grams of Chondrite, natural radio nuclides, fission products such as Ag110m, Cs137 and Cs134 in sea water and marine organism from Pacific Ocean and activated nuclides induced by environmental neutrons and those by fission neutrons (e.g. NAA of Asteroid Itokawa). I will refer to the correction of the cascade sum coincidence.

Log: 243. **OPTIMIZATION OF A MEASUREMENT METHOD WHEN DETERMINING SR-90.** Holmgren, S.(1); Tovedal, A.(1); Björnham, O.(1); Ramebäck, H.(1,2). (1) Swedish Defense Research Agency (FOI), Division of CBRN Defense and Security. (2) Chalmers University of Technology, Department of Chemical and Biological Engineering, Nuclear Chemistry.

When measuring radioactive strontium, after e.g. a reactor accident, the primary radionuclide of interest is the long lived Sr-90 ( $t_{1/2}=28.8$  y). The general aim of this paper is to contribute to a more rapid determination by making the Cherenkov measurement of the daughter nuclide Y-90 more time efficient, with regards to ingrowth as well as measuring time. There are many instances when an optimization of the measurement method might be favorable, such as; situations requiring rapid results in order to make urgent decisions or, on the other hand, to maximize the throughput of samples in a limited available time span. In order to minimize the total analysis time, a mathematical model has been developed which calculates the time of ingrowth as well as individual measuring times for all samples in a series. This work is focused on the measurement of Y-90 during ingrowth, after an initial chemical separation of strontium, in which no other radioactive strontium isotopes are assumed to be present. By using a fixed detection limit and iterating the measurement time for each consecutive sample the total analysis time will decrease, compared to using the same measurement time for all samples. It was found that by optimization, the total analysis time for 40 samples can be decreased from 83 h to 11 h, when assuming a detection limit of 1 Bq/sample and at a background count rate of 0.8 cpm.

Log: 244. **THE BEHAVIOUR OF SELECTED FISSION PRODUCTS AND ACTINIDES ON UTEVA RESIN.** Knight, D. (1); Skinner, M.G. (2); AWE (1,2).

This talk describes the experimental method development at AWE that has led to possible alternative methods for the separation of uranium and plutonium from fission products by extraction chromatography. The behaviour of selected fission product elements and actinides on UTEVA resin was determined by eluting separate columns with decreasing concentrations of nitric acid and hydrochloric acid. A mixed solution of Sr, Y, Zr, Mo, Ag, Cd, Cs, Ba, Ce, Eu, Tb, U, Np and Pu was loaded on to UTEVA resin and eluted with decreasing concentrations of 8M to 0.5M nitric acid. This experiment was repeated with concentrated to 0.03M hydrochloric acid. This initial investigation uses stable solutions for the fission product elements. No additional valency adjustment was employed before separation. The eluted fractions were analysed using ICP-OES to determine the distribution of the majority of the fission products and U; the remaining elements of interest, Cs, Np and Pu were analysed by ICP-MS. The talk will include details of experiments performed to date, chemical yield results, possible applications of the developed work to existing radiochemistry separation schemes at AWE and also planned future validation work. A comparison using valency adjustment on the separation of Pu and Np from other fission products will also be considered. The methods under development have the potential to provide a quick and simple alternative separation of uranium and plutonium from fission products at AWE that could be applied to samples for nuclear forensics purposes. Future validation work includes performing separation methods on radioactive samples.

Log: 245. **RADIOANALYTICAL METHODS: STUDIES OF EFFECTS OF SUPPLEMENTAL N-3 PUFA IN DIET.** Pavelka, S.(1,2); (1) Institute of Physiology, Czech Acad. Sci., Prague. (2) Institute of Biochemistry, Faculty of Science, Masaryk University, Brno, Czech Republic.

As shown in numerous clinical trials and animal experimental studies, n-3 polyunsaturated fatty acids (n-3 PUFA), mostly of marine fish origin, are well known food components recommended for the prevention of cardiovascular diseases, atherosclerosis, and management of hyperlipidemia (for references see e.g. Rauchova H., Vokurkova M., Pavelka S. et al., *Horm. Metab. Res.* 45 (2013) 507-512). The hypolipidemic effect of n-3 PUFA is not completely understood and can be explained by several mechanisms. For example, there are some indications of a potential cross-talk between n-3 PUFA and thyroid hormones in the liver. Therefore, in the present study, we tried to test whether and in what way n-3 PUFA supplementation can affect changes of lipid metabolism, induced in rats by alteration of their thyroid status. In spite of the fact that several biochemical parameters, analyzed with the use of radioanalytical methods, such as total T3 and T4 serum levels, enzyme activity of liver mitochondrial glycerol-3-phosphate dehydrogenase, concentrations of blood glucose, cholesterol, and plasma lipids, were shown to be distinct amongst rats maintained in different thyroid states (euthyroid (EU) vs. hyperthyroid (TH) vs. hypothyroid (HY)), we found no significant effect of 6-week-supplementation of n-3 PUFA on the above mentioned parameters. Thus, n-3 PUFA supplementation did not ameliorate changes induced in the rats by altering their thyroid states.

Log: 246. **RADIOMETRIC ENZYME ASSAYS USED IN STUDIES OF FLUOXETINE INTERACTION WITH THE METABOLISM OF THYROID HORMONES.** Pavelka, S.(1,2); (1) Institute of Physiology, Czech Acad. Sci., Prague. (2) Institute of Biochemistry, Faculty of Science, Masaryk University, Brno, Czech Republic.

Fluoxetine (Fluox) is a representative of a group of non-tricyclic antidepressant drugs known as selective serotonin re-uptake inhibitors, and is the most frequently used drug today for the treatment of episodic depression. Thyroid hormones (TH) can modulate concentration of neurotransmitters and metabolites in the CNS and are supposed to control in this way the activity of some neurotransmitters (e.g., serotonin), which are hypothetically involved in the pathogenesis of depressive illness. One of the pathogenic factors of depression might be, for example, inadequate activities of brain enzymes iodothyronine deiodinases (IDs), which can lead to local insufficient triiodothyronine (T3) concentration. In the present studies, we followed in details the influence of subchronic administration to Wistar rats of Fluox by itself, or in combination with T3, on the metabolism of TH in different rat tissues. To detect changes in the IDs activities in the brain and pituitary, we applied our newly developed radiometric assays for IDs of types 1, 2 and 3. We also employed the adapted radiometric enzyme assays for iodothyronine sulfotransferases (ST) and uridine 5'-diphospho-glucuronyltransferase (UDP-GT) for the assessment of the effects of Fluox on potential induction of activities of these conjugating enzymes in the rat liver. With the use of these radiometric enzyme

assays, we were able to quantify the effects of administered FluoX on the metabolism of TH in the rats. Especially, we determined changes in the most important conjugating and deiodinating enzyme activities in the liver, brain, pituitary and several other tissues of the treated rats.

Log: 247. **RADIOMETRIC QUANTIFICATION: COMPARISON OF GOITROGENIC AND THYROTOXIC EFFECTS OF EXOGENOUS BROMIDE AND PERCHLORATE IONS.** Pavelka, S.(1,2); (1) Institute of Physiology, Czech Acad. Sci., Prague. (2) Institute of Biochemistry, Faculty of Science, Masaryk University, Brno, Czech Republic.

In contrast to iodine, which forms an irreplaceable part of thyroid hormones (TH), in case of similar halogen bromine, there is not enough information available on its biological function and metabolism (for references see e.g. Pavelka S., Chapter 20 in the book: Comprehensive Handbook of Iodine, Acad. Press, Oxford 2009, pp. 199-206). The nature of the toxic effects of bromide on the thyroid gland and the molecular mechanisms of its interference with the biosynthesis of TH has not been explained so far. Goitrogenic effects of bromide may be assumed (Pavelka S., Chapter 61 in the book: Comprehensive Handbook of Iodine, Acad. Press, Oxford 2009, pp. 587-595), similar to those of another inorganic goitrogenic agent perchlorate. However, we have shown recently in the isolated rat thyroids (Pavelka S., J. Radioanal. Nucl. Chem. 291 (2012) 405-408) marked effects of exogenous bromide and perchlorate ions on the activity of the key enzyme in TH metabolism, on thyroid peroxidase (TPO). Here, we followed in more details, with the aid of an improved radiometric enzyme assay for TPO, the influence of an enhanced bromide and/or perchlorate intakes in the animals, maintained on diets with diverse iodine content, on the TPO activity. The use of the adapted radiometric assay revealed a divergent influence of similar goitrogenic agents, bromide and perchlorate ions, on the TPO activity in the rat thyroids. Excessive bromide exerted a biphasic effect, depending on the extent of bromide intake in the animals. The inhibitory effect of bromide was markedly increased in animals maintained on the iodine-deficient diet. In contrast, in all the rats that were administered with high amounts of perchlorate were found elevated TPO activities.

Log: 248. **A NOVEL TECHNIQUE FOR THE RAPID DETERMINATION OF TRIBUTYL PHOSPHATE DEGRADATION FROM ALKALINE HYDROLYSIS IN AQUEOUS AND ORGANIC PHASES USING FTIR-ATR AND VERIFICATION OF THIS TECHNIQUE BY GAS CHROMATOGRAPHY.** Gillens, A.R. (1); Powell, B.P. (1). (1) Clemson University.

Tributyl phosphate (TBP) used in the Plutonium and Uranium Reduction Extraction (PUREX) Process yields degradation products, which makes it inefficient to recover plutonium and uranium after repeated use. Although typically recycled, TBP may be discarded by a caustic treatment with sodium hydroxide, which completely degrades TBP into butanol and sodium salts of dibutyl phosphate (NaDBP). This work describes the development of a novel technique that uses FTIR-ATR measurements of TBP and butanol mixtures to determine the extent of TBP degradation in an alkaline hydrolysis reaction of TBP with 12.5 M sodium hydroxide (NaOH). This technique was verified by analyzing the concentration of TBP and butanol over time using gas chromatography. The FTIR-ATR technique is based on six peak intensity ratios involving peaks that are prominent in the hydrolysis reaction of TBP in 12.5 M NaOH. Linear trends produced by the peak intensity ratios of TBP and butanol mixtures were used to determine the ratios of TBP and butanol in samples retrieved from the hydrolysis reaction. The FTIR study was validated by gas chromatography measurements which quantified the exact concentrations of TBP and degradation products. The amount of TBP in the organic phase decreases from 95% to 36% after approximately four and a half hours in the reaction. The amount of TBP present in the organic phase predicted by the FTIR-ATR technique shows that TBP decreases from 97% to 42% within a similar time frame. The solid phase extraction method that enabled this study utilizes Bond Elut PPL cartridges to recover TBP and DBP that has partitioned into the sodium salt layer that formed from the alkaline hydrolysis of TBP. Up to 29% of TBP and 40% of DBP were extracted from the salt layer based on the extraction recoveries determined. The extraction recoveries of TBP and DBP were also determined for Bond Elut PPL cartridges using nitric acid, water, and sodium nitrate solutions. This paper details a semi-quantitative method for determining TBP degradation from alkaline hydrolysis using FTIR-ATR accompanied by GC verification and it provides a method to extract TBP from aqueous media associated with its use in nuclear reprocessing.

Log: 249. **Multivariable analyses of driving factors of submarine groundwater discharge based on Radon-222 concentrations of the coastal water in Japan.**

Taniguchi, M. (1); Honda, H. (1); Ono, M. (2); Hosono, T. (3); Umezawa, Y. (4); Sugimoto, R. (5); Yamada, M. (1). (1) RIHN. (2) AIST. (3) Kumamoto Univ. (4) Nagasaki Univ. (5) Fukui Pref. Univ.

Radon-222 ( $^{222}\text{Rn}$ ) is one of the useful tracers for evaluating submarine groundwater discharge (SGD), because the groundwater has extremely high concentration in  $^{222}\text{Rn}$  compared with surface water. In this study, we attempted to compile and compare radon data observed in ten study areas in Japan, with SGD observed by seepage meters as well as local environmental parameters such as precipitation, gradient, permeability, basin area, tidal range, and others. We used geographical information system (GIS) and digital elevation model (DEM) to evaluate the characteristics of the basin. The highest  $^{222}\text{Rn}$  concentrations was recorded in Yuza (Northwestern Tohoku area) in Japan, where the highest SGD by seepage meter was observed.  $^{222}\text{Rn}$  concentrations increases with precipitation within the study areas of four bays in Yuza and four bays in Otsuchi. Multivariable analyses of driving factors of submarine groundwater discharge were performed in the ten study areas.

Log: 250. **FUNDAMENTAL AN(III)-LN(III) SEPARATION CHEMISTRY OF N-DONOR LIGANDS.** Geist, A., Karlsruhe Institute of Technology (KIT).

For three decades lipophilic N-donor extracting agents have been used to separate trivalent actinides from the chemically similar lanthanide ions in the context of advanced nuclear fuel cycles. 15 years ago, bis-triazinyl-pyridines (BTP) were shown to achieve this separation under conditions relevant to the reprocessing of nuclear fuels, i.e. from nitric acid solutions. BTP were later followed by the bis-triazinyl-bipyridines (BTBP). More recently, water soluble BT(B)P were used as complexing agents selective for actinides. These could be useful for simplified process schemes. Despite BT(B)P's advantageous properties, current work in international projects is directed towards finding ligands with even better properties. This requires a deep understanding of how such ligands work on the molecular level. This question has been addressed in several studies, the outcome of which has recently been summarised. The following conclusions are drawn: Only N-donor ligands forming strong complexes with actinides(III) show adequate performance in highly acidic solutions. Furthermore, high selectivity over lanthanides(III) seems to correlate with the ligands' ability of forming complexes with exclusive coordination via N atoms. However, several important questions remain unanswered: Why are BT(B)P the only compounds able to extract actinide nitrates from nitric acid with high selectivity, while most other compounds require a more lipophilic anion to form extractable complexes? Is selectivity an intrinsic property of the ligand, or is it induced by its ability of forming complexes with pure N-donor coordination? Can we find even better ligands? Or are BT(B)P the holy grail of An(III)/Ln(III) separation? This presentation discusses the development of N-donor ligands and their properties as selective extracting or complexing agents for actinides(III). Furthermore, it gives an overview of the more fundamental studies performed with BT(B)P (but also with some less efficient N-donor ligands). We show on the one hand how these studies have forwarded our understanding of N-donor-actinide interactions and on the other hand point out some of the open questions to be addressed in the future.

Log: 251. **Direct measurements of long-lived radionuclides in zirconium hulls by LC-ICP/MS.** VIGNEAU Olivier, ARNAL Nadège. CEA, DEN, DEC, SA3C, Laboratoire d'Analyses Radiochimiques et Chimiques, F13108 Saint Paul lez Durance, FRANCE.

The measurement of long-lived radionuclides (LLR) is required in applications of nuclear concern (such as nuclearpower reactors operating and waste management). Radionuclides are usually present at trace or ultratrace levels so extraction step and chemical treatments are often required before measurements. These are often tedious, require significant amounts of radioactive samples leading to exposure of operators to substantial doses and cause problems for waste management. By coupling a liquid chromatograph and an ICP/MS, it is possible to measure low levels of LLR in complex matrix such as zirconium hulls (high-level solid waste) of spent fuel assemblies. The on-line separation of the components of the sample eliminates many stages of chemical treatments, apart from the dissolution of the solid sample and the dilution of the obtained solution. In addition, the volumes handled are greatly reduced, decreasing the absorbed dose for operators. Finally, the nature and the volume of generated waste are easily manageable by the current sector. Ion exchange resins enable the separation of the LLR of interest from the interfering stable elements or polyatomic ones. The LC-ICP/MS technique is applied to analyze LLR such as  $^{135}\text{Cs}$  and  $^{99}\text{Tc}$  and other radionuclides (such as  $^{137}\text{Cs}$ ) at ppb and ppt levels contained in zirconium hulls, which may content Sn, Nb, Mo, Ru and Ba. The accuracy and precision of the method is demonstrated by comparison of the amount content of  $^{137}\text{Cs}$  of the zirconium hulls determined by LC-ICP/MS with the one measured by gamma spectrometry.

Log: 252. **Accurate and precise  $^{235}\text{U}$  quantification by combining isotopic dilution method and ICP/AES measurements.** VIGNEAU Olivier, ARNAL Nadège. CEA, DEN, DEC, SA3C, Laboratoire d'Analyses Radiochimiques et Chimiques, F13108 Saint Paul lez Durance, FRANCE.

An ICP/AES owning a high resolving power is able to measure the isotopic shift in emission atomic lines for heavy elements such as uranium. Uranium isotopes, such  $^{235}\text{U}$  and  $^{238}\text{U}$ , are therefore clearly separated and identified. Using this phenomenon, the accurate and precise quantification of uranium or its isotopes is possible. Indeed, the transposition of isotopic dilution mass spectrometry method to our ICP/AES instrument allows to quantify, with trueness and precision, the amount content of  $^{235}\text{U}$  of an uranyl nitrate solution. The method is named IDAES. At our knowledge, this study is the first that combine isotope dilution method and ICP/AES measurements. In order to demonstrate the validity of the IDAES method, an IDMS protocol (the preferred method), using a MC ICP/MS instrument, is also applied to determine the amount content of  $^{235}\text{U}$  of the sample. Both methods led to the same amount content of  $^{235}\text{U}$  for the uranyl nitrate solution (difference between both values is less than 0.1%). As expected, the precision of the IDMS method (less than 1%) is better than the IDAES one (about 3%) but it remains satisfactory. Those results are particularly remarkable since IDAES and IDMS used physically different measuring systems, respectively ICP/AES and MC ICP/MS.



Log: 253. **IMPROVED GAMMA-SPECTROSCOPY OF MARINE SAMPLES VIA LOW BACKGROUND SMALL ANODE GERMANIUM (SAGE) WELL DETECTOR.** Pike, S.M.(1); Adekola, A.(2); Colaresi, J.(2); Ilie, G.(2); Mueller, W.F.(2); Yocum, K.M.(2); Buesseler, K.O.(1). (1) Woods Hole Oceanographic Institution. (2) CANBERRA Industries, Inc.

Analysis of marine and environmental samples by gamma-spectroscopy is non-destructive and multiple isotopes can be identified from a single spectrum. In 2013 Woods Hole Oceanographic Institution (WHOI) was invited to be a beta test site for CANBERRA Industries next generation of germanium well detectors. The enhanced energy resolution and efficiency of the Small Anode Germanium (SAGE) well detector was combined with a cosmic-veto suppression system and used to count marine sediments and water samples. To evaluate the performance of the SAGE, several different types of marine samples were compared to existing traditional high purity germanium well detectors (HPGe). The new cosmic-veto suppression system reduced the shielded background of the SAGE by an average of 35% across the energy spectrum with negligible full-energy peak losses. In comparative measurements for dating of marine sediments from off the coast of Japan, the improved energy resolution of the SAGE was able to separate major U-238 decay chain energies 92, 238 and 241 keV  $\gamma$  rays which traditional HPGe well detectors could not. In measurements of cesium isotopes in seawater extracted onto an absorber, the SAGE was able to separate the 134 cesium peak at 604 keV  $\gamma$  ray from the 609 keV  $\gamma$  line of Bi-214. The reduced background combined with improved energy resolution and efficiencies of the SAGE well detector with cosmic-veto suppression system and lower detection limits has shortened counting times and increased sample through put. Results from the study of natural and Fukushima derived radionuclides in marine samples will be presented.

Log: 254. **IS THERE STILL A SPACE FOR "HOMEMADE" SEPARATION MATERIALS FOR DEDICATED SEPARATIONS AND RADIOANALYSIS?.** Němec, M. Czech Technical University in Prague.

In the field of analytical separation methods, especially those used in nuclear and radiochemistry, the mainstream methods are nowadays clearly divided into two diverging streams. In the first one, the separation methods or even the complete detailed separation procedures are designed – sometimes even “bended” – to fit the commercially available rather general-purpose materials. In the second stream, the approach is opposite – materials with application-specific properties are designed and developed. The Department of Nuclear Chemistry at CTU in Prague (DNC) continues its long tradition in the development and preparation of novel separation materials and alternative separation procedures. “Homemade” materials are widely produced and used for a broad scale of specific separation tasks. The properties of such materials are fine-tuned to meet requirements of their foreseen application. The key factors in the development of an “ideal” (radio)analytical separation method are not only a high separation specificity but also providing optimum/simple conditions for the following measurements of the analyte and determination of the separation yield. Such policy is then leading to the limitation of the number of separation steps or sometimes even to a merger of the separation and

measurement. Selected examples of separation materials and methods recently designed or under development at the DNC will be presented e.g. preparation and testing of clean separation materials, separations combined with extractive LSC measurement, or a system for Am separation from NPP evaporator concentrate.

Log: 255. **PRECONCENTRATION OF TRACE URANIUM FROM IRON-RICH MATRICES USING DIPEX EXTRACTANT.** Trauth, A.M. (1); Lavelle, K. (1); Glover, S.E. (2); Connick, W.B. (1); Spitz, H.B. (1). (1) University of Cincinnati. (2) NIOSH.

A preconcentration method using Eichrom Technologies' DIPEX® Actinide Resin has been developed for isotopic measurement of trace quantities of actinides, such as uranium or plutonium, in an iron rich sample matrix. Iron-bearing materials are known to locally concentrate uranium in ground and surface waters and have been used as environmental preconcentrators of uranium and plutonium. Typically, large quantities of sample are required to reliably detect long lived, alpha-emitting radioactive isotopes with low specific activity. Unfortunately, large samples will include interferences that can increase background and adversely affect detection limits. This paper describes a method suitable for analyzing trace quantities of actinides in environmental samples that eliminates the matrix interference by removing >98% of iron in solutions containing up to 1.0 g iron per 3 mL resin.

Log: 256. **A COMPARISON OF TECHNIQUES FOR THE DETERMINATION OF NEPTUNIUM IN PLUTONIUM.** Thompson P.L; Kaye P.; Huggins A.N; Danbury R.; King J.

237 neptunium is a common radiogenic impurity of plutonium produced by the beta decay of 241 plutonium to 241americium and the subsequent alpha decay of the americium. Np 237 concentration is of interest in the fields of plutonium metallurgy and age dating. A number of analytical methods have been trialled for the separation and determination of Np 237 from a plutonium matrix. These include a new direct measurement without separation using high resolution Inductively Coupled Plasma Mass Spectrometry (ICPMS), an isotope dilution separation using TEVA resin coupled with High Resolution Gamma Spectrometry (HRGS) and a strong base anion exchange method also coupled with HRGS. The advantages and disadvantages of each technique and the accuracy and precision achieved are discussed.

Log: 257. **A chemical system for rutherfordium using TEHA and TEHP.** Jeff Rolfes (1), John Despotopulos (1,2), Roger Henderson(2), Dawn Shaughnessy(2), Ralf Sudowe(1) (1)University of Nevada, Las Vegas; (2)Lawrence Livermore National Laboratory.

Studies of the chemical properties of the heaviest elements have always been difficult due to the short half-lives and low cross sections involved. To solve this problem, atom-at-a-time methods are necessary to determine the properties of short-lived isotopes. Extremely fast kinetics for the chemical reactions studied (on the same order as the nuclide's half-life) are required, and the system should have potential for automation. One potential technique, extraction chromatography, offers a way of investigating rutherfordium's properties, without the solvent waste

generated in liquid-liquid extractions. A rapid method involving the use of a ligand with high intragroup selectivity is necessary to discover more about element 104. Studies with the homologs of rutherfordium, zirconium and hafnium, can be performed using liquid-liquid extractions as well as extraction chromatography. From literature reviews, tertiary amines and phosphates show promise in extracting group 4 elements. In particular, tris(2-ethylhexyl)amine (TEHA) and tris(2-ethylhexyl)phosphate(TEHP) can be used to selectively extract zirconium and hafnium from a chloride matrix. Results of a study investigating the extraction of Zr and Hf from various mineral acids by these two ligands will be presented. The development of an extraction system for rutherfordium based on these extractants can help to further elucidate its properties.

Log: 258. **PRODUCTION OF ACTIVATION SPECIES FROM HIGH-ENERGY NEUTRON ( $E_n \geq 20$  MEV) IRRADIATIONS.** Bandong, B.B.(1); Roberts, K.E.(1); Harward, N.K. (1); Haslett, R.J.(1); Wang, T.F.(1); Padgett, S.W.(1); Hartman, S.C.(2). (1) Lawrence Livermore National Laboratory. (2) University of California, Davis.

In support of research and development for nuclear forensics, we have been investigating the production of activation species by irradiating elemental foils of Al, Ti, Fe, Ni, and Au with high-energy neutrons ( $E_n \geq 20$  MeV) produced by the reaction of 30- to 40-MeV deuterons on a thick beryllium target at the Crocker Nuclear Laboratory of the University of California at Davis. The neutron spectrum peaks at around 20 MeV and extends up to 40 MeV, producing a distribution of neutron-deficient nuclides by various (n,xn) reactions and neutron-rich nuclides via (n,gamma), (n,p) and (n,alpha) reactions. We will describe our compact beryllium target and activation foils holder assembly and present the observed neutron flux and spectrum at the position of the foils. We will present production pathways and rates of activation species observed from the irradiation of Al, Ti, Fe, Ni, and Au elemental foils with high-energy neutrons and discuss challenges associated with their production and quantitation via high-resolution singles gamma-ray spectrometry. Geant4 simulation was also performed to characterize the neutron flux out of the Be sputtering target and to guide future improvements in the Be target – activation foil assembly.

Log: 259. **DISPERSION AND EXPORT DYNAMICS OF  $^{137}\text{Cs}$  DEPOSITED ON THE FORESTED ECOSYSTEM IN FUKUSHIMA AFTER THE NUCLEAR POWER PLANT ACCIDENT IN MARCH 2011.** Ohte, N.(1); Murakami, M.(2); Endo, I.(3); Iseda, K.(3); Tanoi, K.(3); Ishii, N.(4). (1) Kyoto University. (2) Chiba University. (3) University of Tokyo. (4) National Institute of Radiological Sciences.

The accident of the Fukushima Daiichi Nuclear Power Stations in March 2011 emitted  $1.2\text{E}16\text{Bq}$  of  $^{137}\text{Cs}$  into the surrounding environment. Radioactive substances including  $^{137}\text{Cs}$  deposited onto the forest areas of the northeastern region of Japan. Forests of these regions are especially important for forest production industry, but also for the source areas of drinking water. The first phase of the governmental survey and investigations showed that major portion of deposited  $^{137}\text{Cs}$  is trapped on the canopies and litter layers of soil surface. As  $^{137}\text{Cs}$  is easily adsorbed onto clay minerals in soil, major portion of those can be transported by eroded soils and particulate organic matters with water discharge. Dissolved  $^{137}\text{Cs}$  can be taken up by microbes, algae and plants in soil and aquatic systems. Then,  $^{137}\text{Cs}$  are eventually introduced into insects, worms, fishes and birds through the food web. To clarify the mechanisms of dispersion and export of  $^{137}\text{Cs}$  within and from a forested ecosystem, we have conducted intensive observations and samplings in a forested headwater catchment in the area ~50 km distant from the Nuclear Power Station. Expected two major pathways of  $^{137}\text{Cs}$  transportations are; 1) by water movement via dissolved and particulate or colloidal forms, 2) dispersion through the food web in the forest-stream ecological continuum. The  $^{137}\text{Cs}$  concentrations of stream waters are monitored. Various aquatic and terrestrial organisms are periodically sampled to measure their  $^{137}\text{Cs}$  concentrations. Following findings have been derived: 1) Major forms of exported  $^{137}\text{Cs}$  are suspended matters. Especially, particulate organic matters may be most important carrier of  $^{137}\text{Cs}$ . High flow generated by a storm event accelerate the transportation of  $^{137}\text{Cs}$  by water flows from forested catchments. Estimation of  $^{137}\text{Cs}$  export from the forested catchments requires precise evaluation of the high flow acceleration during storm events. 2) Because the biggest pool of  $^{137}\text{Cs}$  in the forested ecosystem is accumulation of litters and their detritus, the  $^{137}\text{Cs}$  dispersion is quicker through the detritus food chain than the grazing food chain. Especially in aquatic systems, most predators have already taken  $^{137}\text{Cs}$ . An urgent question is when and how the  $^{137}\text{Cs}$  disperse through the grazing food chains, and how quick that process will occur. To solve or predict these phenomena, mechanisms of  $^{137}\text{Cs}$  releasing from litters and soil organic matter should be clarified.

Log: 260. **PHENYL TRIFLUOROMETHYL SULFONE AS DILUENT IN A GROUPED ACTINIDE EXTRACTION PROCESS.** Halleröd, J.; Engdahl, E.; Ekberg, E.; Aneheim, E. Chalmers University of Technology .

At Chalmers university of technology, the main research focus within the transmutation and separation field has been the development of a Grouped ActiNide EXtraction (GANEX) process. The main idea with partitioning and transmutation is to separate the transuranic elements and the fission products in used nuclear fuel from each other and then use a fast neutron spectrum to transmute the transuranic elements. The GANEX process is based on two cycles; in the first cycle the uranium bulk is removed from the fuel dissolution liquor and in the second cycle (the actual GANEX extraction is performed in this step) the transuranic elements and the remaining uranium are extracted together to avoid pure plutonium streams. A GANEX process that combines the two extractants 6,6'-bis(5,6-dialkyl-[1,2,4-triazin-3-yl])-2,2'-bipyridine (CyMe4-BTBP) and tri-butyl phosphate (TBP) in cyclohexanone has previously been developed at Chalmers. However, due to the low flash point (44 C) of cyclohexanone, the high water solubility and its degradation in contact with the acidic aqueous phase it is desirable to replace cyclohexanone with a different diluent. It is, however, not easy to find a proper diluent to the GANEX process based on CyMe4-BTBP and TBP. CyMe4-BTBP is hard to dissolve in aliphatic diluents and the extraction kinetics is slow in diluents such as alcohols and aromatics. Therefore another alternative has to be found. Recently phenyl trifluoromethyl sulfone (FS-13) has been investigated as diluent. Due to stability against irradiation and degradation, a high flashpoint (122 C), solvents with 70 % FS-13, 30 % TBP and 10 mM CyMe4-BTBP has been investigated.

Log: 261. **REACTOR SOURCE TERM DISCRIMINATION USING ENVIRONMENTAL CESIUM ISOTOPE RATIO MEASUREMENTS: NUCLEAR FORENSIC APPLICATIONS.** Snow, M.S.(1); Snyder, D.C.(1). (1) Idaho National Laboratory.

Cs isotope ratio analyses provide a valuable, emerging tool for nuclear forensic investigations; as several isotopes of Cs (e.g. Cs-134, Cs-135, and Cs-137) are produced in varying quantities during nuclear fission, Cs isotope analyses have the potential to provide critical nuclear forensic information such as reactor neutronics (including energies, fluxes and reactor duty cycles) and age constraints for a given fission event. Environmental samples represent an abundant yet challenging source of nuclear forensic samples; Cs isotope analyses from such samples are complicated by low Cs abundances and the necessity for purification from isobaric and ionization inhibiting interferences which are present at extremely high concentrations relative to fission product Cs. In this work, we present Cs isotope analyses from environmental samples as a forensic tool for environmental source term discrimination. A detailed discussion of the major challenges which currently confront these analyses will be presented. The ultimate ability for source term discrimination will be demonstrated from soil and vegetation samples taken several hundred kilometers from the Fukushima Daiichi site, where the combination of actinide and Cs data enables discrimination of the source of the environmental contamination among a variety of different source terms (including nuclear weapons fallout, the Unit 1-3 reactor cores, and Unit 1-4 spent fuel cooling ponds). Results from Cs and actinide analyses from locations around the world will also be presented along with the potential application of Cs isotope analyses for reactor source term discrimination in future studies.

Log: 262. **HIGHLY-SENSITIVE AMS AND COINCIDENCE/ANTICOINCIDENCE HPGe SPECTROMETRY ANALYSIS OF RADIONUCLIDES IN MARTIAN METEORITES.** Povinec, P.P.(1); Sýkora, I.(1); KOváčik, A.(1); Jull, A.J.T.(2); Caffee, M.(3); Koeberl, C.(4,5); Feriere, F.(5); Brandstaetter, F.(5). (1) Comenius University in Bratislava. (2) University of Arizona at Tucson. (3) Purdue University. (4) University of Vienna. (5) Natural History Museum in Vienna.

Cosmogenic (e.g.,  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{60}\text{Co}$ ) and primordial (e.g.,  $^{40}\text{K}$ ,  $^{232}\text{Th}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ) radionuclides in meteorites have been useful tracers for a better understanding of their pre-atmospheric dimensions, cosmic-ray exposure ages, and terrestrial ages. Their low concentrations in meteorites require highly sensitive analytical methods, such as accelerator mass spectrometry (AMS) for analysis of long-lived radionuclides, or high sensitive gamma-spectrometry for analysis of gamma-ray emitters. While AMS analyses require radiochemical pre-treatment of samples and preparation of suitable targets for production of ion beams from analyzed elements, gamma-spectrometric analyses of meteorite fragments can be carried out non-destructively. Specific analytical challenges, such as AMS analysis of radionuclides in sub-gram meteorite samples, or efficiency calibration of HPGe detectors for non-destructive analysis of large meteorite fragments, will be discussed in detail. Examples of applications of both methods for the analysis of Martian meteorites will be presented, and the obtained results will be used for a better characterization of their origin as well as their cosmic and terrestrial ages.

Log: 263. **Neutron Activation Imaging to Locate Hot Particles.** Kurth, M. (1); Cutright, J. (1); Glover, S.E.(2); Spitz, H.B.(2); Cao, L.R.(1). (1) The Ohio State University. (2) University of Cincinnati.

Clusters of micrometer- or nanometer-sized radioactive material, termed hot particles, contain the majority of radioactivity released in the nuclear fuel cycle, in accident scenarios and also provide means to determine origin and processes used in nefarious activities. The analysis of these particles can lead to isotope content, environmental transport, health effects, and safeguards to proliferation. A current issue is the lengthy investigative times needed to locate particles commonly suspended in a matrix of soil and/or on a filter paper. While large plutonium particles (~50 micrometers) of weapons grade plutonium have both alpha and gamma activity high enough to allow for several methods of identification and isolation, particles of enriched and highly enriched uranium are much more difficult to detect unless they have a fission component (from a reactor or a fission device). A neutron radiography device with a custom configured phosphor screen is used to detect hot particles. Phosphor screen loaded with boron compound is used to cancel background as well as to provide quantitative information about particles' radioactivity level. A thermal neutron beam at the Ohio State Research Reactor is used to activate the fissile hot particles inducing fission fragments and subsequent scintillation. This novel active interrogation method is capable of delivering accurate locations of hot particles within a matrix in fast measurement times. The method was evaluated using weapons grade plutonium and highly enriched uranium particles that had been characterized using scanning electron microscopy.

Log: 264. **NUCLEAR SCIENCE AND TECHNOLOGY IN ACHIEVING SUSTAINABLE AGRICULTURE IN DEVELOPING COUNTRIES.** Tanzania Atomic Energy Agency.

Tanzania Development Vision TDV 2025, which aims to develop a nation with middle income status by the year 2025, has three goals; Attaining a high quality livelihood, Attaining good governance and rule of law, and Building a strong and competitive economy. In attaining a high quality livelihood goal, the target for food self sufficiency and food security rank in the first out of nine set targets. This is because of its important contribution to national economic growth, noting that 95% feed its population of more than 44.9 million, 74% of the country's labor force is employed in the agricultural sector, the sector is contributing to more than 28% of the Gross Domestic Product, foreign currency through export earnings and more than 65% as input for local industries. This sector is currently characterized by low production, poor quality products, and low return. Low production and poor quality are products of unfavorable conditions resulting from climate change, pests, soil fertility, and water scarcity for agricultural production. Greater productivity, food safety, high quality products and environmental sustainability in the agricultural sector can be substantially achieved through the application of nuclear science and technology. Properly applied, this will increase food production, generate the surpluses that will be used in industries for export, increase employment as well as improve food safety—thereby moving Tanzania to middle income country status by 2025. This paper outlines the current contributions of nuclear science and technologies in the agriculture sector in different parts of the world and the challenges that need to be addressed to allow nuclear science to help achieve sustainable agriculture development in United Republic of Tanzania to eradicate poverty, farmers & pastoralist conflicts, and food related diseases.



Log: 265. **URANIUM ELECTROCHEMICAL DETECTION AND ANALYSIS IN LITHIUM CHLORIDE-POTASSIUM CHLORIDE EUTECTIC AS AN ASSESSMENT FOR MATERIAL ACCOUNTABILITY IN PYROPROCESSING TECHNOLOGY.** Phongikaroon, S. (1); Hoover, R.O. (2). (1) Virginia Commonwealth University. (2) University of Idaho.

Pyroprocessing technology, currently operated at the Idaho National Laboratory (originally developed by Argonne National Laboratory), is a dry separation method that can be applied to used nuclear fuel from the Experimental Breeder Reactor-II. The main component of this process is the Mark-IV electrorefiner (ER), which is used to electrochemically oxidize and dissolve the uranium from anode baskets into the electrolyte and then reduce it to metal at the solid steel cathode. Few experimental studies have been done in the past with uranium at concentrations approaching those conditions occurring in the Mark-IV ER. Thus, a set of experiments including cyclic voltammetry (CV), chronopotentiometry (CP), and anodic stripping voltammetry (ASV), has been conducted to explore and provide an assessment for advancing material accountability toward this important process. Uranium experiments were performed at 773 K with concentrations from 1.0 to 10.0 wt% uranium chloride in the lithium chloride-potassium chloride eutectic molten salt. From these experiments, diffusivity, apparent standard reduction potential and activity coefficient can be calculated. Further results will be presented and discussed.

Log: 266. **RAPID QUANTITATION OF URANIUM FROM MIXED FISSION PRODUCT SAMPLES.** Seiner, B.N. (1); Eggemeyer, T.A. (1); Beck, C.L. (1); Finn, E.C. (1); Friese, J.I. (1). (1) Pacific Northwest National Laboratory.

Rapid determination of uranium isotope activity from nuclear accidents is useful for emergency response and environmental assessment. However, measurement by radiological detection methods such as gamma spectroscopy and alpha spectroscopy can be challenging due to the presence of other gamma and alpha emitting fission and activation products. The detection limit of the methods can be improved through a robust chemical separation of uranium from these other fission products prior to analysis. The purpose of this work was to establish an improved and more rapid chemical separation of uranium from a mixed fission product sample using multiple Eichrom® UTEVA cartridges. Due to the large fraction of fission product tellurium (Te) co-eluting through the UTEVA with the U, a subsequent spontaneous deposition of Te on Ag method was developed. The optimized method demonstrated a consistent chemical yield of  $74 \pm 3$  % for uranium. This procedure was evaluated using fission products from an irradiated HEU sample. The uranium was chemically yielded by KPA, and the minimum detectable activity (MDA) determined following HPGE gamma spectroscopy. From this sample no detectable activity of gamma emitting fission products was measured and the MDA for U-235 and U-238 was reduced by a factor of 1137 and 1113, respectively. The chemical isolation of uranium was successfully achieved in less than four hours.

Log: 267. **MEASUREMENT OF RELATIVE FISSION PRODUCT YIELDS FROM 12.2 MEV DEUTERON-INDUCED FISSION OF PU-240.** Judah Frieese; Lori Metz; Erin Finn; Larry Greenwood; Stephanie Gregory; Leah Arrigo; Rick Wittman. Pacific Northwest National Laboratory.

Measurements of deuteron-induced fission are rare. In this work, a sample containing fission products from 12.2 MeV-induced fission of highly isotopically enriched Pu-240 was available as a byproduct of another experiment. The non-volatile fission products were initially quantified by singles gamma spectrometry on the dissolved target. The number of fissions was estimated at  $3.13\text{E}11$  ( $\pm 19.8\%$ ) using fast pooled neutron-induced fission yield data for peak fission products. Further chemical separations were performed to isolate fission products not quantified by the bulk count. The fission product distribution observed is distinct from that typically observed for neutron-induced fission and lacks the usual double-humped profile. However, it is similar to fission product distributions from other non-neutron induced fission work. Fission product yields reported here are relative as the cross-section for fission under the conditions for this sample has not been reported and this experiment did not measure the absolute fission rate.

Log: 268. **FISSION PRODUCTS MEASURED FROM HIGHLY-ENRICHED URANIUM IRRADIATED UNDER 10B4C IN A RESEARCH REACTOR.** Lori Metz (1); Erin Finn (1); Larry Greenwood (1); Judah Frieese (1); Corey Hines (2); Mat King (2); Don Wall (2). (1) Pacific Northwest National Laboratory. (2) Washington State University.

Critical assemblies provide one method of achieving a fast neutron spectrum that is close to a U-235 fission-energy neutron spectrum for nuclear data measurements. Previous work has demonstrated the use of a natural boron carbide capsule for spectral-tailoring in a mixed spectrum reactor as an alternate and complementary method for performing fission-energy neutron experiments. Previous fission products measurements showed that the neutron spectrum achievable with natural boron carbide was not as hard as what can be achieved with critical assemblies. New measurements performed with the WSU TRIGA reactor using a boron carbide capsule 96% enriched in 10B for irradiations resulted in a neutron spectrum very similar to a critical assembly and a pure U-235 fission spectrum. The current work describes an experiment involving a highly-enriched uranium target irradiated under the new 10B4C capsule. Fission product yields were measured following radiochemical separations and are presented here. Reactor dosimetry measurements for characterizing neutron spectra and fluence for the enriched boron carbide capsule and critical assemblies are also discussed.

Log: 269. **ISOTOPIC ANALYSIS OF PLUTONIUM IMPURITY IN NEPTUNIUM TARGET.** Noyes, K.L. (1); Gregory, S.J. (1); Haney, M.M. (1); and Lucas, D.D. (1). (1) Pacific Northwest National Laboratory.

Recently a Np-237 target (26 mg) was analyzed for trace Pu isotopes (approximately 300 ppb). This work describes the chemistry required for a Np/Pu separation sufficient for Thermal Ionization Mass Spectrometry (TIMS), as well as the Pu purification from other potential contaminants. A point source was prepared from the purified Pu fraction and the atom ratios were measured via TIMS. The Pu isotopics provide a picture of the Np target's irradiation history, which will also be discussed.

Log: 270. **SUITABILITY OF BCR-482 AS A LICHEN REFERENCE MATERIAL FOR TRACE ENVIRONMENTAL RADIONUCLIDE MEASUREMENTS.** Lavelle, K.B.(1); Miller, J.L. (2); Hanson, S.K.(2); Connick, W.B.(1); Spitz, H.B.(1); Glover, S.E.(1); and Oldham, W.J.(2). (1) University of Cincinnati (2) Los Alamos National Laboratory.

There is no widely available lichen material that has been well characterized for anthropogenic radionuclides, despite the considerable and long-standing interest in the use of lichen for monitoring of environmental radioactivity. Existing lichen reference materials designed for use in analytical laboratories engaged in monitoring air quality are certified for the content of heavy metals. In this work, select anthropogenic radionuclides have been measured in the commercially available reference material BCR 482 in order to evaluate whether reproducible measurements suitable for interlaboratory comparisons may be obtained for these analytes. BCR 482 was collected in Axalp, Switzerland in 1991 and is composed of the epiphytic lichen *Pseudevernia furfuracea*. Samples taken from three separate bottles of BCR 482 were analyzed for uranium, neptunium, and plutonium isotopes by inductively coupled plasma mass spectrometry (ICP-MS) and analyzed for cesium-137 by gamma-ray spectrometry. The measured results were consistent between samples for all analytes, suggesting that BCR 482 is a suitable material for comparisons of these types of measurements. Further confidence in the results is gained due to the fact that the isotopic composition of the sample is consistent with the expectation that anthropogenic radionuclides present in the lichen are primarily composed of a mixture of global fallout and the more volatile materials released following the Chernobyl accident.

**Log: 271. DEVELOPMENT OF AMMONIUM BIFLUORIDE FUSION METHOD FOR RAPID DISSOLUTION OF TRINITITE SAMPLES AND ANALYSIS BY ICP-MS.**

Brockman, J. D.(1); Hubley, N.(2); Brown, J. W.(3) Robertson, J. D. (1) (3). (1) University of Missouri Research Reactor. (2) Hope College. (3) University of Missouri-Columbia.

Rapid analysis of radionuclides in environmental samples is important for nuclear forensics programs. There is a need to develop fast, field deployable dissolution techniques that are compatible with existing instrumental techniques for rapid radionuclide analysis. Recently, fusion with ammonium bifluoride ( $\text{NH}_4\text{HF}_2$ ) has been demonstrated to dissolve refractory, felsic, and ultramafic minerals. The solid salt  $\text{NH}_4\text{HF}_2$  decomposes at 125 °C to form HF vapor capable of breaking Si-O bonds. This study examined the capability of fusion with  $\text{NH}_4\text{HF}_2$  to dissolve trinitite. The trinitite sample was ground to a powder using a ball mill. A 50 mg sample of trinitite was loaded into a PFA vial with 250 mg of  $\text{NH}_4\text{HF}_2$  and heated at 230 °C for 3 hours. The residue was dissolved in 8 M nitric acid, heated to dryness, and re-dissolved in 2% nitric acid. The solution was filtered using 1  $\mu\text{m}$  filter paper with no residue observed. The  $\text{NH}_4\text{HF}_2$  fusion process was evaluated by comparison with digestion blanks and conventional microwave digestion. A second sample was dissolved using HF and  $\text{HNO}_3$  acids in a sealed PTFE vessel using a Milestone Ethos plus microwave digestion station. High resolution inductively coupled plasma mass spectrometry was used to measure the concentration of 33 elements in the trinitite samples prepared by both  $\text{NH}_4\text{HF}_2$  and microwave digestion.

**Log: 272. APPLICATION OF PLUTONIUM AND CESIUM ISOTOPE RATIOS TO ESTIMATE THE DURATION OF IRRADIATION OF SPENT REACTOR FUEL.**

Robel, M. (1); Ramon, E. C. (1); Isselhardt, B. H. (1); Bleuel, D. L. (1); Conant, A. J. (1); Erickson, A. (2); and Carney, K. P. (3). (1) Lawrence Livermore National Laboratory. (2) Georgia Institute of Technology. (3) Idaho National Laboratory.

In this work, we describe a novel application of actinide and fission product isotope ratio measurements to the estimation of the duration of irradiation of spent fuel samples. This period of irradiation monitor (PIM) utilizes the time independent correlation of the Cs-135/Cs-137 isotope ratio to local neutron flux, in combination with the Pu-240/Pu-239 fluence monitor to calculate the length of time a sample of spent fuel was irradiated. This investigation includes both modeling and experimental validation using both gamma and mass spectrometry analysis of samples of spent fuel from the BR3 reactor. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory (under contract DE-AC52-07NA27344). LLNL-ABS-665175

Log: 273. **Radiation Effects on Li-ion Battery in Rescuing Robots at Post-nuclear Accident.** Tan, C.T. (1); Lyons, D.(2); Co, A.(2); Cao, L.R.(1). (1) Department of Nuclear Engineering, The Ohio State University. (2) Department of Chemistry and Biochemistry, The Ohio State University.

Rechargeable li-ion batteries (LIBs) are now widely used as power source in industry, households and scientific research. It is also used as the power source in vehicles for outer space exploration and in robots for performing rescue, sampling missions in highly radioactive environments such as at post-nuclear accident. The intense radiation environment may degrade the battery quickly to the state of non-operation or reduce the system's performance. In this paper, we studies the radiation effects on LIBs' capacity and some critical performance parameters when placed in high gamma dose environment. Coin cells of LIBs were assembled and tested after the electrodes, electrolyte and its components were irradiated using a  $^{60}\text{Co}$  irradiator at the Nuclear Reactor Lab of The Ohio State University under a fluence ranging from hundreds of krad/h to Mard/h. Each component was irradiated before assembling into the cell and the battery capacity as well as the electrical performance was characterized.

Log: 274. **Theoretical Investigation of Isotope Exchange Reaction in Tritium-Contaminated Mineral Oil in Vacuum Pump.** Liang Dong (1), Liang Du (1), Zhao-Yi Tan (1). (1) Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics.

The mechanism of the isotope exchange reaction between tritium and several typical organic molecules in vacuum pump mineral oil has been investigated by density functional theory (DFT), and the reaction rates are determined by conventional transition state theory (TST). The tritium-hydrogen isotope exchange reaction is accomplished through one hydrogen migration step to produce the tritiated product. Isotope exchange between hydrogen and tritium is selective, and oil containing molecules with  $-\text{OH}$  and  $-\text{COOH}$  groups can more easily exchange hydrogen for tritium. For aldehydes and ketones, the isotope exchange ability is determined by the hydrogen on the alkyl group rather than that on the oxygen-containing group. The molecules containing one type of hydrogen provide a single product, while the molecules containing different types of hydrogens provide competitive products. The rate constants are presented to quantitatively estimate the selectivity of the products.

Log: 275. **Characterization of Boron Carbide Based Polymer Neutron Sensor with Neutron Depth Profiling.** Tan, C.T. (1); Dong B. (2); James R. (2); Kelber J. (2); Turkoglu D. (1); Downing R.G. (3); Cao L.R. (1). (1) The Ohio State University. (2) University of North Texas. (3) National Institute of Standards and Technology.

Neutron is a unique signature emitted from actinides that can be detected for safeguarding special nuclear materials for enhancing homeland security. Fabrication of diode detectors based on novel boron carbide: aromatic composites has recently been developed at University of North Texas. These new materials exhibit enhanced charge collection and neutron voltaic performance compared to BC films. The new materials contain both carboranes (B<sub>10</sub>C<sub>2</sub>H<sub>12</sub>) and aromatic species, and were deposited by plasma enhanced chemical vapor deposition (PECVD). Since low Z elements in these films are typical difficult to measure by conventional method (e.g., X-ray), a neutron beam based analytical technology termed as neutron depth profiling (NDP) was used to determine the BC and BC: aromatic film thickness. Samples were characterized at the cold neutron NDP facility at U.S. National Institute of Standards and Technology. The total film thickness as well as the concentration distribution of B were determined for the calibration deposition rates during PECVD and provide accurate information for neutron deflection efficiency and capacitance measurements. This paper will mainly focus on the demonstration of NDP as a unique analytical method for characterizing thin film neutron detectors.

Log: 276. **ANALYSIS OF FOOD MONITORING DATA BEFORE AND AFTER THE FUKUSHIMA NUCLEAR ACCIDENT.** Steinhauser, G.(1); Shozugawa, K.(2); Merz, S. (3). (1) Colorado State University. (2) University of Tokyo. (3) Vienna University of Technology.

In an unprecedented food monitoring campaign for radionuclides, the Japanese government took action to secure food safety after the Fukushima nuclear accident (11 March 2011). We analyzed a part of the immense data set, in particular radiocesium contaminations in food from the first year after the accident. Activity concentrations in vegetables peaked immediately after the campaign had commenced, but they decreased quickly, so that by early summer 2011 only few samples exceeded the regulatory limits. Later, accumulating mushrooms and dried produce led to several exceedances of the limits again. Monitoring of meat started with significant delay, especially outside Fukushima prefecture. After a buildup period, contamination levels of meat peaked by July 2011 (beef). Levels then decreased quickly, but peaked again in September 2011, which was primarily due to boar and deer meat (both known accumulators of radiocesium). Pre-Fukushima Cs-137 and Sr-90 levels (resulting from atmospheric nuclear explosions) in food were typically lower than 0.5 Bq/kg, whereby meat was typically higher in Cs-137, and vegetarian produce was usually higher in Sr-90.

Log: 277. **REDUCING UNCERTAINTIES FOR SHORT LIVED FISSION PRODUCT YIELDS.** Sean Stave, Amanda Prinke, Larry Greenwood, Derek Haas (1); Jason T. Burke, Jennifer Jo Ressler, Anton P. Tonchev, Walid Younes (2). (1) Pacific Northwest National Laboratory. (2) Lawrence Livermore National Laboratory.

Fission product yields are important for applications such as heat production in nuclear energy generation, nuclear waste product inventories, and radiopharmacology. The uncertainties associated with short lived (half-lives less than one day) fission product yields listed in databases such as the National Nuclear Data Center's ENDF/B-VII or the Nuclear Energy Agency's JEFF are large enough for certain isotopes to provide an opportunity for new precision measurements to offer significant uncertainty reductions. Many uncertainties for short lived fission products are based upon model estimates and not experimental data. A series of experiments has begun where small samples of U-235, U-238, and Pu-239 are irradiated with a pulsed, fission neutron spectrum at the Nevada National Security Site. The irradiated samples are then placed between two 65% relative efficiency broad-energy germanium detectors. Both single and coincident gamma-ray data are collected in list mode spanning the time from approximately 1 hour to 1 week post-irradiation. Both the energy and decay time of the resulting gamma-ray signatures are used to identify the fission products present in the whole sample. The amount of various isotopes present immediately following the irradiation can be determined given the total counts and the calibrated properties of the detector system. Data analysis has begun and the uncertainty on the fission yields for multiple isotopes has been reduced by nearly an order of magnitude. The experimental methods and results will be reviewed in this paper. This work was funded by the Office of Defense Nuclear Nonproliferation Research and Development within the U.S. Department of Energy's National Nuclear Security Administration. This document is PNNL-SA-106962.

Log: 278. **Environmentally friendly sample preparation methods for uranium oxide dissolution.** L Meyers, T Yoshida, N Xu Los Alamos National Laboratory Chemistry Division.

There is a need to develop green sample preparation methods that can carry out the dissolution of uranium oxide on swipes for in-field characterization. Ammonium bifluoride (ABF,  $\text{NH}_4\text{HF}_2$ ) has been used as a dissolution reagent for a variety of compounds, including beryllium oxide. Carbonates in the presence of an oxidant (e.g.  $\text{NaOCl}$  or  $\text{H}_2\text{O}_2$ ) have originally been used for uranium metal and alloy dissolutions as well as uranium oxides. It has been discovered through various ABF and  $\text{CO}_3 + \text{H}_2\text{O}_2$  experiments that these two methods are selective at dissolving uranium oxides. Bulk uranium oxide dissolution techniques were first explored and then applied to various swipe media.  $\text{U}_3\text{O}_8$  is almost completely dissolved by ABF in an hour, while  $\text{UO}_2$  is fully dissolved by  $\text{CO}_3 + \text{H}_2\text{O}_2$  in 10 minutes.  $\text{UO}_2$  partially dissolves in the presence of ABF and so does  $\text{U}_3\text{O}_8$  when added to a  $\text{CO}_3 + \text{H}_2\text{O}_2$  mixture. The chemicals included in the reaction are green (i.e. environmental friendly) and less hazardous compared to strong mineral acids that are currently most commonly used; therefore, they can be handled and transported during an in-field application easily. LA-UR-14-29027.

Log: 279. **Measuring the photon depth dose distribution produced by a medical linear accelerator in a water-equivalent radio-fluorogenic gel.** Sandwall, P.A. (1); Spitz, H.B. (2), Elson, H.R. (2); Lamba, M.A.S. (2); Connick, W.B. (2); Fenichel, H. (2). (1) Vantage Oncology (2) University of Cincinnati.

Radio-fluorogenesis is the formation of fluorescent products in response to ionizing radiation. This study describes use of a radio-fluorogenic gel to obtain quality assurance measurements for a mega-voltage photon beam from a medical linear accelerator. Dose deposition in water from ionizing radiation results in radiolysis, yielding hydroxyl free radicals. In aqueous solution, coumarin-3-carboxylic acid undergoes hydroxylation to form 7-hydroxy-coumarin-3-carboxylic acid, a derivative of umbelliferone. A known fluorescent probe, umbelliferone exhibits peak excitation in UV to near UV light (365-405nm) with a visible 445nm emission. Instrumental analysis enables determination of absorbed dose by quantification of fluorescent emissions. Depth dose distributions were obtained with a radio-fluorogenic gel comprised of aqueous gelatin and coumarin-3-carboxylic acid. Compared with vendor published values, the measured clinically relevant uncertainty was less than 3% in nearly all areas of electronic equilibrium. This is similar to results from other clinical dosimeters, such as metal-oxide field effect transistors (MOSFET) and thermo-luminescent dosimeters (TLD), which report uncertainties near 7%. With additional investigations to characterize uncertainty in areas of non-electronic equilibrium, this research suggests radio-fluorogenic gel dosimeters could be used for quality assurance of clinical photon beams.



Log: 280. **PERFORMANCE OF A LOW ACTIVITY BETA-SENSITIVE SR90 WATER MONITOR FOR FUKUSHIMA.** Zickefoose, J.(1); Bronson, F.(1); Ilie, G.(1); Jaderstrom, H (1); Venkataraman, R.(1). (1) CANBERRA Industries.

There are large volumes of contaminated water from the stabilization efforts at the damaged Fukushima Nuclear Power Plants. This water is being processed to remove radioactivity for eventual release to the environment. An on-line continuously operating system to confirm that the clean-up system is working properly, and to provide prompt feedback of the results is required by the system operator. While gamma emitting nuclides allow for the straight forward approach of gamma spectroscopy to identify and quantify radioactivity in water, pure beta emitting nuclides such as Sr90 pose a challenging problem. The relatively short range of beta radiation in water requires optimization of the measurement geometry in terms of the source-detector distance and source-detector interface while retaining a background sensitivity low enough to meet the Minimum Detectable Concentration (MDC) of 10 Bq/kg in 180 minutes. The use of the Monte-Carlo code MCNP to estimate system performance before prototyping vastly increases the success of the end product. Various parameters such as detector size and thickness, water chamber size, water chamber construction materials were evaluated to help choose the optimum geometry. The final design was a system consisting of two large-area (16x35cm) and thin (0.15mm) plastic scintillators placed very close to a sealed water chamber. The size of the chamber was optimized to obtain the maximum efficiency for the nuclide being measured (Sr/Y90) but to minimize the efficiency for possible interferences (Ru/Rh106, Cs137). A thin carbon fiber window was selected with adequate material and thickness to contain the water under pressure, but also thin enough (0.5mm) to allow enough beta radiation to pass through to the active detector volume. The entire measurement geometry is then housed in a thick lead shield to reduce contributions from external sources to an acceptable level. Data acquisition is accomplished through customized application-specific software that allows for long counting times to attain a low MDC, but also simultaneously provides alarms on short averaging times to achieve a fast response to sudden changes in activity concentration. Multiple monitors are then linked to supervisory software where real time data and alarms are available remotely. Testing of the production devices has shown that we can achieve the 10 Bq/kg MDC requirement for Sr90 in equilibrium with Y90 with a count time of approximately 20 minutes.

Log: 281. **NEW TECHNOLOGY FOR ISOTOPIC RATIO MEASUREMENTS WITHOUT A MASS SPECTROMETER: LAMIS (LASER ABLATION MOLECULAR ISOTOPIC SPECTROSCOPY).** Russo, R.E. (1,2); Mao, X.L. (1); Chan, GC (1); Bol'shakov, A (2); Yoo, J.(2). (1) Lawrence Berkeley National Laboratory. (2) Applied Spectra, Inc.

Traditionally, laser ablation (LA) sampling has been coupled with ICP-MS (Inductively Coupled Plasma Mass Spectrometry) for sensitive isotopic analysis. Alternatively, optical emission from the luminous plasma initiated by the ablation process at the sample surface can be monitored and provide elemental analysis and in some cases isotopic analysis. This well-known optical approach is called LIBS (Laser-Induced Breakdown Spectroscopy). Overall, LIBS is not ideally suited for isotopic analysis due to poorly resolved spectral shifts in atomic and ionic spectra from isotopes. However, our new technology LAMIS (Laser Ablation Molecular Isotopic Spectroscopy) shows the ability to perform isotope measurements in these laser plasmas at atmospheric pressure. By expanding the capabilities of classical LIBS to emphasize the measurement of molecular emission spectra in addition to elemental, LAMIS provides the ability to measure all elements and their isotopes, especially light elements like Li, Be, C, N, O which are impossible with XRF. We developed LAMIS to date by demonstrating its ability to measure B, C, H, D, Sr and other isotopes. We demonstrated low percent levels for sensitivity and have experimental plans to meet ppm levels. For some isotopes, we have achieved < 0.1% precision. The talk will describe the isotope work that has been reported in LIBS plasmas and show how LAMIS expands those capabilities.

Log: 282. **Effect of radiation and *Schewanella putrefaciens* on the sorption behavior of U(VI) on goethite.** Lee, J.K.; Lee, S.Y.; Jeong, J.; Baik, M.H., Korea Atomic Energy Research Institute (KAERI).

In a deep geological environment, the mobility of radionuclides released from a high-level radioactive waste (HLW) repository can be highly affected by the surrounding environment such as the groundwater, microbial activity, and radiation. In this study, batch type experiments were carried out to investigate the influence of  $\gamma$ -irradiation and iron reducing bacteria on the sorption of U(VI) on goethite for 36 days in an anoxic condition. *Schewanella putrefaciens* (*S. putrefaciens*) was used as the iron reducing bacteria. Samples were irradiated for 5 days at a dose rate of 0 and 2 Gy/hr for a total dose of 0 and 230 Gy using a Co-60 gamma source. The effect of  $\gamma$ -irradiation on the sorption of U(VI) on goethite was higher than that of *S. putrefaciens*. When the sample was irradiated, the sorption amount was increased by about 20% regardless of the microbes while it was slightly increased in the presence of *S. putrefaciens*. Though it has little effect on the sorption, *S. putrefaciens* considerably changes the solution chemistry. The redox potential of irradiated sample in the presence of *S. putrefaciens* significantly decreased to  $-28 \pm 10$  mV compared with that of non-irradiated sample of  $+462 \pm 16$  mV. The ionizing radiation and microbial activity induced the dissolution of iron from goethite as Fe(II) to form a magnetite-like mineral, which might facilitate the reduction of U(VI) to uranium (IV). Moreover, the radiation at a total dose of 230 Gy had no effect on the death and regrowth of *S. putrefaciens*.

Log: 283. **REDUCTION OF PU(VI) IN ACIDIC SOLUTIONS WITH ORDERED MESOPOROUS CARBON MICROSPHERE-PACKED COLUMNS.** Parsons-Moss, T. (1,2); Uribe, E. (2,3); Wu, Z.(4); Wang, J.(5); Zhao, D.(5). (1) Lawrence Livermore National Laboratory, Glenn T Seaborg Institute. (2) Nuclear Science Division, Lawrence Berkeley National Laboratory. (3) Department of Chemistry, University of California, Berkeley. (4) Department of Chemistry, Soochow University. (5) Department of Chemistry and Laboratory of Advanced Materials, Fudan University. Most laboratory processes involving separation, recovery or purification of plutonium rely on manipulation of the Pu oxidation state. Electrochemical reduction of Pu(VI) can be achieved with chemical reducing agents, leading to further purification steps, or with a potentiostat, which can be very time-consuming. We recently showed with batch contact experiments and optical absorbance spectroscopy that Pu(VI) in 1 to 1.5 M acid solutions (HCl, HClO<sub>4</sub>, and HNO<sub>3</sub>) is spontaneously reduced to lower oxidation states by contact with high-surface-area porous carbon powders. The extent and rate of reduction depends on the solution conditions, and the surface area and surface chemistry of the carbon. It was demonstrated with amorphous activated carbon powder that Pu(VI) in 1.2 M HClO<sub>4</sub> solution is reduced to Pu(III) when pushed through a glass column packed with porous carbon particles. Ordered mesoporous carbon (OMC) hollow microspheres were synthesized to optimize these columns. Advantages of the OMC spheres include high specific surface area, and regularity in both particle and pore size and shape. The capacity of this material for reduction of Pu(VI) was explored, and found to be higher than that of activated carbon. Column parameters and conditions were investigated to optimize the OMC column for quick and reliable reduction of Pu(VI) to Pu(III). Spectroscopic measurements of the carbon surfaces before and after the redox reaction were made to determine which species were oxidized. The reducing property of porous carbon beads may be a useful tool for plutonium chemists, and could potentially be combined with other resins or extraction agents in novel separation schemes.

Log: 284. **THERMAL ANALYSIS OF AUSTRALIAN URANIUM ORE CONCENTRATES FOR MATERIALS PROVENANCING IN NUCLEAR FORENSIC SCIENCE.** Ditcham, T. (1); Wotherspoon, A. (2); Kirkbride, P. (1); Lenehan, C. (1); Popelka-Filcoff, R.S. (1). (1) School of Chemical and Physical Sciences, Flinders University. (2) Australian Nuclear Science and Technology Organisation.

Identifying the provenance of illicitly trafficked nuclear material is an important facet of nuclear forensic science, with 2477 confirmed incidents reported between 1995-2013 [1]. Uranium ore concentrates (UOCs) are a material of particular interest to nuclear forensic science as they contain greater than 65% uranium by weight and are a commodity traded internationally for the production of nuclear fuel. Unfortunately, the current capabilities of nuclear forensic science results in the inconclusive determination of an unknown UOC's origin, necessitating the development of new analytical techniques and signatures in UOCs to remove the ambiguity observed in provenance studies. Research described in this presentation revolves around the novel use of thermogravimetric analysis (TGA) and differential scanning calorimetry, in concert with micro-Raman spectroscopy, X-ray diffraction and scanning electron microscopy, to examine the physicochemical properties of Australian UOC samples at elevated temperatures. Triuranium octoxide (U<sub>3</sub>O<sub>8</sub>) samples from Ranger, NT and Olympic Dam, SA can be readily distinguished from uranium peroxide (UO<sub>4</sub>.xH<sub>2</sub>O) samples from Beverley, SA through TGA and micro-Raman spectroscopy, as U<sub>3</sub>O<sub>8</sub> is thermally stable at elevated temperatures [2], whereas UO<sub>4</sub>.xH<sub>2</sub>O decomposes to U<sub>3</sub>O<sub>8</sub>, through a number of uranium trioxide (UO<sub>3</sub>) intermediates [3]. TGA, complimented with scanning electron microscopy, X-ray diffraction and micro-Raman spectroscopy, has demonstrated the capability to differentiate between different species of UOC compounds originating from a number of Australian uranium mines. These preliminary results suggest that thermal analysis of UOCs may contribute in a nuclear forensic science investigation, to assist in the determination of unknown sample's provenance.

Log: 285. **SPECIATION ANALYSIS OF IODINE ISOTOPES (127I AND 129I) IN AEROSOL USING SEQUENTIAL EXTRACTION AND MASS SPECTROMETRY TECHNIQUES.** Luyuan Zhang (1); Xiaolin Hou (1,2), Sheng Xu (3), Yukun Fan (2), Weijian Zhou (2) (1) Center for Nuclear Technologies, Technical University of Denmark, Risø Campus, Roskilde 4000, Denmark. (2) Xi'an AMS Center, SKLLQG, Institute of Earth Environment, CAS, Xi'an 710075, China. (3) Scottish Universities Environmental Research Center, East Kilbride, G75 0QF, UK.

A method has been developed to analyze species of 127I and 129I in aerosol samples. The aerosol sample was collected using a polypropylene filter with pore size of 0.45 µm which was attached to an in-house aerosol sampler at Risø campus, Technical University of Denmark. Air flux of the sampler is about 1500 m<sup>3</sup> h<sup>-1</sup>. A portion of aerosol filter was cut to small pieces and extracted sequentially with water and sodium hydroxide solution. Total iodine and non-extractable iodine on the filter was separated from filter matrix by alkaline ashing method. Iodide and iodate in the water leachate was separated using anion exchange chromatography, and these iodine fractions were further separated with solvent extraction, and finally prepared to AgI for measurement of 129I using accelerator mass spectrometry. 127I in each fraction was measured by ICP-MS. The parameters for separation of each species of iodine including leaching time and temperature, alkaline ashing condition were investigated in detail, and an optimal analytical procedure was established. The method has been applied for analysis of a time series aerosol samples collecting in Denmark, the results showed that iodide is the major specie of water soluble iodine, and organic iodine is the predominant form of total iodine in aerosol.

**Log: 286. Development of new strategies for measurement of Cl-36 in nuclear steel samples at very low level by using AMS technique.** R. Brennetot (1), P. Perret (1), C. Colin (1), P. Fichet (1), E. Laporte (1), R. Haudebourg (1), C. Andrieu (2), F. Goutelard (3)(1) CEA/DEN/DANS/DPC/SEARS/LASE, CEA Saclay, Bat 459 PC 171, 91191 Gif sur Yvette, France(2) Electricite de France, EDF – CIDEN / Departement Etudes - Division Deconstruction/Groupe Inventaire et Agrements, 154 Avenue Thiers, CS 60018, 69458 LYON CEDEX 06(3) CEA/DEN/MAR/DEIM/Nuclab, CEA Marcoule Bat 109 BP 17 171, 30207 Bagnols-sur-Ceze, France.

Waste management is a key issue for nuclear industry. In France, ANDRA (the national agency for nuclear waste management) is in charge to size, build and operate waste repositories. In order to assure the security of these facilities, a list of radionuclides and their maximum associated activities has been defined as acceptance criteria by ANDRA. From this list, some are readily and easily measured while many, such as pure beta emitters are difficult to measure, requiring long radiochemical processes. Due to its high mobility and long half-life, the acceptance limit for Cl-36 is as low as 5 Bq/g, making it as one of the radio-isotope that will size the final repository of the nuclear waste considered. In several cases, result of the measurements carried out, after radiochemical process, with Liquid Scintillation Counting (LSC) detection technique is under the detection limit of typically 1 Bq/g. In the lack of other data, the detection limit is automatically considered by the stakeholder to overestimate the waste activity. Overestimated waste activity could dramatically increase the cost and will reduce the radioactive capacity of the storage facilities. To overcome this problem, the Operator Support Analyses Laboratory has developed new radioanalytical procedures to measure the radionuclide of interest (Cl-36, Be-10) by Accelerator Mass Spectrometry (AMS) at level as low as 1 mBq/g in nuclear wastes matrixes. Two procedures dedicated to Cl-36 measurement have been developed for activated steel samples. The first one, a simplified radiochemical method has been implemented in order to analyze Cl-36 with a detection limit around 1Bq/g by LSC. A second procedure has been developed to prepare sample for AMS measurements. Radiochemical blank and homemade standard have been analyzed to calibrate the analytical procedure. In the particular case of contaminated steels the analytical process has been reviewed including Digital Autoradiography (DA) before and after acid attack of the samples. DA technique is a nondestructive analytical technique sensitive to all types of radionuclides providing images of radioactivity. Cl-36 images recorded on steel samples have allowed to prove the efficiency of the sample preparation and also the sample homogeneity. The limit of detection for contaminated samples will be discussed

Log: 287. **Measurements of long-lived Sn-121m and Sn-126 nuclides in Low and Intermediate Level Nuclear Wastes.** Gautier C. (1), Coppo M. (1), Degros J.P. (1), Fichet P. (1), Goutelard F. (2). (1) Operator Support Analyses Laboratory, Atomic Energy Commission, CEA Saclay, DEN/DANS/DPC/SEARS/LASE, Building 459, PC171, 91191 Gif-sur-Yvette Cedex, FRANCE. (2) CEA/DEN/MAR/DEIM/Nuclab, Atomic Energy Commission, CEA Marcoule, Building 109, BP 17 171, 30207 Bagnols-sur-Ceze, FRANCE.

In France, nuclear wastes are managed by the National Radioactive Waste Management Agency (ANDRA). Several repository sites have been built in order to accommodate nuclear waste packages. One is dedicated to wastes containing short-lived radionuclides with radioactivity at Low and Intermediate Level. Criteria for 143 radionuclides have been defined by ANDRA which guarantees the safety of the facility. Among this long list, Sn-121m and Sn-126 have to be declared as soon as their activities are over 10E-3 Bq/g. Both tin isotopes are fission products obtained according to thermal fission yields of 0.00003 % and 0.06 %, respectively. Sn-121m can also be produced by activation of tin present as an additive in zircalloys and as an impurity in inconels and steels used in the nuclear industry. Sn-121m decays with a half-life of 55 years whereas Sn-126 is a very long-lived radionuclide with a half-life estimated at around 10E5 years. Because of the long half-life and the unknown impact of high energy gamma emissions of Sn-126, ANDRA allows a very low Sn-126 activity limit (2.7 Bq/g) in waste packages. Due to their potential low activities in nuclear wastes, both tin nuclides need to be separated from the matrices and concentrated through chemical separations prior to any measurement. From a literature review, it can be emphasized that only a few radiochemical procedures have been published. So, the LASE (Operator Support Analyses Laboratory) laboratory of CEA Saclay (France) has developed a selective radiochemical method for their measurements in nuclear wastes. The procedure starts with a microwave digestion or alkaline fusion with the addition of a natural tin carrier and a 113Sn radioactive spiker. The following steps are constituted of ion exchange separations combined with co-precipitations and a highly selective ethylation step. The final purified fractions thus obtained are suitable for gamma-spectrometry analysis of Sn-121m and Q-ICPMS measurements of Sn-126. The protocol yield is evaluated by gamma measurement of 113Sn. The overall separation yield varies from 45 % to 70 % depending on matrix, with a detection limit of about 20 Bq/g for Sn-121m and 0.1 Bq/g for Sn-126, respectively. The presentation will first focus on the optimization and the validation of the procedure. The second part of the work will be addressed to its application to various types of intermediate level nuclear matrices (activated steels and ion-exchange resins).

Log: 288. **SPARSE-VIEW NEUTRON CT RECONSTRUCTION OF IRRADIATED FUEL ASSEMBLY USING TOTAL VARIATION MINIMIZATION WITH POISSON STATISTICS.** Muhammad Abir (1); Fahima Islam (1); Daniel Wachs (2); Hyoungh Koo Lee (1). (1) Department of Mining and Nuclear Engineering, Missouri University of Science and Technology, Rolla, MO 65409. (2) Idaho National Laboratory, 2525 N Fremont Ave, Idaho Falls, ID 83402.

The investigation of irradiated nuclear fuel assembly can be crucial for analyzing coolant flow through the channels and for the identification of structural changes of the assembly due to radiation damage. We present an exemplary study of the fuel assembly inspection demonstrating the potential use of sparse-view neutron CT in this field. Neutron projection images of the fuel assembly are collected at the Idaho National Laboratory (INL) hot cell facility using indirect foil-film transfer technique. The radiographs are digitized using a commercial film digitizer and registered spatially for reconstruction. Digitized data are reconstructed using simultaneous iterative reconstruction technique (SIRT) with total variation (TV) minimization using dual approach for numerical solution assuming the projection data are corrupted by Poisson noise. The dual approach avoids smoothing of sharp edges, hence preserves jumps in the reconstruction as well as it smooths out the noisy fluctuations. To validate and evaluate the performance of the algorithm, visual inspections as well as quantitative evaluation studies using a computer simulation projection data and the experimental digitized data of the fuel assembly are carried out. The proposed method provides better reconstruction for both simulated and experimental case in terms of artifact reduction, higher SNR, and better spatial resolution compared to the reconstruction yielded by filtered back projection (FBP) and SIRT reconstruction. Keywords: CT reconstruction, sparse-view, total variation minimization, Poisson statistics, irradiated fuel assembly

Log: 289. **PREPARATION OF RADIOXENON AND RADIOARGON MIXED SOURCES FOR IFE 2014.** Biegalski, S.R. (1); Tipping, T.N. (1); Klingberg, F.J. (1). (1) The University of Texas at Austin.

The Integrated Field Exercise (IFE14) was conducted from November 3, 2014 through December 9, 2014 in Jordan. This was a large exercise to test the capabilities for On-Site Inspection (OSI) as part of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). One important OSI technology is the measurement of noble gasses that could be produced within a nuclear explosion. To support IFE14, radioxenon samples and radioargon samples were produced at The University of Texas at Austin. The goal was to produce a mixed sample containing  $^{131}\text{mXe}$ ,  $^{133}\text{Xe}$ , and  $^{37}\text{Ar}$  in activity ratios consistent with the nuclear explosion scenario defined for the exercise. Due to shipping times and the uncertainty in the time required for clearance through Jordanian customs, samples had to be produced well in advance of the start of IFE14. This work describes the production process and the analysis of the samples prior to shipment.



Log: 290. **FISSION PRODUCT ANALYSIS OF HEU IRRADIATED UNDER A FAST-POOLED NEUTRON SPECTRUM: COMPARISON OF DETECTION METHODOLOGIES AT PNNL AND AWE.** Seiner, B.N. (1); Finn, E.C. (1); Greenwood, L.R. (1); Metz, L.A. (1); Friese, J.I. (1); Marsden, O. (2); Davies, A.V. (2); King, A.R. (2); Scivier, P. (2). (1) Pacific Northwest National Laboratory. (2) Atomic Weapons Establishment.

The fission products from HEU formed under a fast pooled neutron spectrum generated using a boron carbide shield were analyzed at Pacific Northwest National Laboratory (PNNL) and the Atomic Weapons Establishment (AWE). At PNNL, the peak fission products were measured from the primary solution using standard HPGe gamma spectroscopy and the low fission yield products (Ag-111, Cd115, and Cs-136/137) were chemically separated prior to HPGe gamma spectroscopy. At AWE, the peak fission products and the select low fission yield products were determined using a Compton suppression gamma spectrometer in addition to the standard HPGe gamma spectroscopy analysis. The results from both laboratories are compared, with significant attention paid to the potential benefits and limitations of the different gamma spectroscopy methods.

Log: 291. **ELECTROCHEMICAL SEPARATION OF URANIUM FROM URANIUM/NEODYMIUM MIXTURE IN IONIC LIQUIDS.** Droessler, J.E.(1); Boland, K.S.(2); Scott, B.L.(2); Hatchett, D.W.(1); Czerwinski, K.R.(1); Runde, W.H.(2); Goff, G.S.(2). (1) University of Nevada Las Vegas, Chemistry Department. (2) Los Alamos National Laboratory.

Both actinides and lanthanides are extremely electropositive and typically require rather large reduction potentials which cannot be accessed in aqueous solution without competing solvent reactions. However, sufficiently negative potentials can be reached in certain ionic liquids (ILs) which can be capitalized upon for the reduction of f-elements at the electrode surface. This provides a basis for the controlled, potential mediated, separation of f-elements by electrodeposition from ILs. This work was an initial investigation of three ILs containing the bis(trifluoromethanesulfonyl)imide anion in regards to electrodeposition of neodymium (Nd) and uranium (U), especially as a mixed sample. Separation of U from Nd by electrodeposition from the ILs was attempted. UV-Visible (UV-Vis) spectroscopy was also used following extraction of the U and Nd from IL samples by contacting with 1 M HClO<sub>4</sub>. This provided the starting relative ratio of U:Nd in the IL samples. Potentiostatic electrodepositions were then conducted on aliquots of the IL samples at various negative potentials (-1.2 V, -2.0 V, and -2.8 V) using Cu foil. Deposits obtained from electrodeposition were cleaned, dried, and then dissolved for UV-Vis analysis of the U/Nd ratio. Spectroscopy of dissolved deposits indicated separation of U from Nd has occurred. Concentrations will be confirmed by ICP-AES analysis. Other analysis conducted on the deposits included scanning electron microscopy (SEM) with energy dispersive x-ray spectroscopy (EDS) and powder X-ray diffraction (p-XRD).

Log: 292. **PUREX Process Signatures for Nuclear Forensic Analysis.** Braley, J.C.(1); Koehl, M.A.(1); Rundberg, R.S. (1) Colorado School of Mines (2) Los Alamos National Laboratory.

Fission product yield data sets are one of the most important and fundamental compilations of basic information in the nuclear industry. This data has a wide range of applications which include nuclear fuel burnup and nonproliferation safeguards. Current data sets present a wide range of uncertainty and demands for improved accuracy of fission product data are prevalent throughout the industry. Relative fission yields constitute a major fraction of the reported yield data and reduce the number of required absolute measurements. R-values are a double ratio of fission product yields where the numerator is the ratio of a given fission product yield to a standard isotope from a given fissile fuel and neutron energy. The denominator is the ratio of the same two fission products in the thermal fission of  $^{235}\text{U}$ . R-values are sensitive to the neutron environment of the fissile fuel and particularly useful when separation methods are used. Radiochemical separations of fission products reduce interferences, facilitate the measurement of low level radionuclides, and are instrumental in the analysis of low-yielding symmetrical fission products. It is especially useful in the measurement of the valley nuclides and those on the extreme wings of the mass yield curve, including lanthanides, where absolute yields have high errors. Early measurements have been made with thermalized neutrons within sampling positions in the US Geological Survey TRIGA Reactor (GSTR) at the Denver Federal Center in Lakewood, CO. These studies have focused on identifying sources of error in radioanalytical separations and detection and removing or reducing them in order to measure relative fission product yield with minimal error.

Log: 293. **DEVELOPMENT OF HIGH ENERGY (BENCH TOP) METHODS FOR THE PREPARATION OF SURROGATE NUCLEAR EXPLOSION DEBRIS (SNED).** Carman, A.J.(1); Liezers, M.(1); Endres, M.C.(1), Eiden, G.C.(1). (Pacific Northwest National Laboratory).

Trinitite has become a favorite material for nuclear forensics studies in part because of its availability as a bona fide example of solid debris resulting from a nuclear detonation. Its bulk composition of fused arkosic desert sand does not adequately represent the type of debris that might form if a nuclear device was detonated in an urban setting where concrete, brick, glass, asphalt, steel, aggregate and stone would form the principle components of the resulting debris. To study the debris that might form in this kind of setting we have been exploring various bench top high energy techniques that could produce materials with some of the characteristics of nuclear explosion debris. The methods under investigation include using inductively coupled plasmas, exploding wires and high power laser heating. The application of these methods will be described along with some of the materials that can be produced with these methods and their similarities to real nuclear explosion debris.

Log: 295. **A NEW METHOD FOR ANALYZING FIRST ROW TRANSITION METALS AND ACTINIDES IN MELT GLASS.** Roman, A.R. (1); Bond, E.M.(1). (1) Los Alamos National Laboratory.

Federal and international agencies rely on quickly obtained and highly accurate technical data for nuclear forensic investigations. This requires rapid and accurate analysis of a variety of samples and matrices. To advance the analytical timeline of samples, the more rapid and efficient a separation can be made the shorter the total time from sample acquisition to data analysis. Many current techniques used are lengthy and complicated. In order to achieve a more simplified and quicker separation, DGA resin was used in the development and investigation of the separation of stable and activated first row transition metals and actinides of interest from a synthetic melt glass. These metals are of interest due to the prompt fast neutron activation produced during a detonation which can provide information about the device. The activation of the first row transition metals were performed by a collaboration of multiple national laboratories. The synthetic melt glass used was produced by University of Nevada, Las Vegas. This separation's efficiency, recoveries, and the length of procedure will be discussed. The fractions from these columns were analyzed by ICP-AES and gamma spectroscopy. 'LA-UR-14-29338'

Log: 296. **PREPARATION OF THIN IRIIDIUM TARGETS BY ELECTRODEPOSITION FOR NEUTRON CAPTURE CROSS SECTION MEASUREMENTS.** Bond, E. M. (1); Moody, W. A. (1); Arnold, C (1); Bredeweg, T. A. (1); Jandel, M. (1); Rusev, G (1). (1) Los Alamos National Laboratory.

The preparation of Ir-191 and Ir-193 electrodeposits for neutron capture cross section measurements at the Detector for Advanced Neutron Capture Experiments (DANCE) located at the at Los Alamos Neutron Science Center (LANSCE) is described. The electrodeposition of iridium in the desired thickness of 0.4-1 mg/cm<sup>2</sup> proved to be challenging. Electrodepositions from ammonium sulfate and isopropyl alcohol solutions were initially attempted with natural iridium spiked with Ir-192; better yields and thicknesses were obtained using molecular plating from isopropyl alcohol solutions. Ir-191 and Ir-193 targets were originally prepared using the standard single-sided electrodeposition cell; however, thicker targets were desired. Therefore, iridium electrodepositions using our double-sided electrodeposition cell were developed. The iridium electrodepositions from the double-sided cell were optimized by varying the time, current, acid concentration and iridium concentration. LA-UR-14-29283

Log: 297. **ALPHA SPECTROSCOPY AND MASS-BASED SEPARATIONS USING ICP-MS.** Dion, M.P. (1); Eiden, G.C. (1), Farmer, O.T. (1); Liezers, M. (1); Robinson, J.R. (1). (1) Pacific Northwest National Laboratory.

The intrinsic mass separation capability of an inductively coupled plasma mass spectrometer has been used to create ion-implanted samples for alpha spectroscopy. We present results on energy resolution improvements measured with Am-241 implanted samples and also evaluate a sample of implanted mass 238 from a certified isotopic standard of Pu-244 by alpha spectroscopy. The results of the 238 (i.e., Pu-238) implant are compared to an electrodeposited sample of the Pu-244 standard. The Am-241 implants attain the intrinsic resolution of the silicon detector used in the measurement implying an exceptionally clean, thin sample. When this concept was applied to a multi-isotope standard (Pu-244, Pu-242, Pu-240, Pu-239, Pu-238 and Am-241), mass 238 was separated to a level 1 part in  $159 \pm 27$  based on alpha activity.

Log: 298. **EVALUATION OF ENVIRONMENTAL AVAILABILITY OF ACTINIDES FROM POST-REMEDIATION PARTICLES ORIGINATING FROM THE 1960 BOMARC INCIDENT.** Bowen, J.M. (1,2); Glover, S.E. (1); Pratt, S.H. (2); Hart, G.L. (2); Spitz, H.B. (1); LaMont, S.P. (3). (1) University of Cincinnati. (2) Pacific Northwest National Laboratory. (3) Los Alamos National Laboratory.

The relationship between the environmental availability of individual actinide-rich particles and their morphological characteristics was examined using a sequential extraction protocol to determine the persistence of nuclear forensic signatures. Five well-characterized plutonium metal particles were extracted from samples of post-remediated soil that had been contaminated by the 1960 BOMARC incident. These particles, which remained in contact with soil and exposed to environmental conditions for over 50 years, were classified as either amorphous or crystalline based upon surface characteristics and appearances observed via scanning electron microscopy. A sequential extraction procedure was used to determine the quantity of plutonium, americium, and uranium in each particle that dissolved after exposure to a series of increasingly aggressive reagents. The results demonstrated that the quantity of actinides extracted was related to the morphological classification so that amorphous particles were likely to be more affected by environmental conditions than crystalline particles. Additionally, the uranium isotopic ratios were determined via inductively coupled plasma mass spectrometry for two select particles, one amorphous and one crystalline, to examine the persistence of enrichment levels throughout the sequential extraction protocol. Since particle morphology affected particle dissolution in reagents simulating environmental conditions, it may be possible to predict the long-term fate of actinide-bearing particles in soil based upon the morphological category of the particle.

Log: 299. **CHARACTERIZATION AND FABRICATION OF A PU TARGET FOR PRODUCTION OF AM-240 VIA DEUTERON BOMBARDMENT.** Bowen, J.M. (1); Finn, E.C. (1); McNamara, B.K. (1); Wittman, R.S. (1). Pacific Northwest National Laboratory.

Many short-lived radioisotopes have applications in radiochemical research and development. However, they are often not commercially available due to their short half-life and must be made on an as-needed basis. Our interest in using Americium-240 (Am-240), produced from the deuteron bombardment of a plutonium-240 (Pu-240) target, necessitated the development of a novel target preparation method. Previous irradiation trials on an electrodeposited Pu target containing ~2 mg of Pu resulted in damage due to heat buildup or flaking of the electrodeposited Pu oxide, reducing the yield of the irradiation. This study evaluates different target preparation techniques with an emphasis on producing a Pu target that is robust to heat and shock and is compatible with post-irradiation radiochemical separations to recover Am-240. A Pu target was produced and characterized for uniformity and robustness using published methods. The improved Pu target will be bombarded with deuterons in an irradiation trial.

Log: 300. **AN ALPHA-CONVERSION ELECTRON COINCIDENT SYSTEM FOR ACTINIDE ISOTOPICS.** Dion, M.P. (1); Miller, B.M. (1), Warren, G.A. (1). (1) Pacific Northwest National Laboratory.

The alpha-conversion electron signature is a unique characteristic of several important actinides. We are pursuing this measurement technique to reduce the chemistry (and time) required for quantification of a sample that contains actinides which exhibit alpha energy overlap. We present the conversion electron spectrum of Am-241, Pu-238 and Pu-239 using a silicon drift detector and compare the results of relative internal conversion coefficients to theoretical values. Furthermore, a coincident measurement device has been assembled with off-the-shelf components. Coincident data from Am-241, Pu-238 and Pu-239 will be presented and a multi-strip silicon detector coincident system will be discussed.

Log: 301. **Characterization of Gallium Nitride for Radiation Detection.** Mulligan, P.L. (1); Cao L.R. (1). (1) The Ohio State University.

The need for room temperature, radiation hard, solid-state radiation detectors has led to the exploration of many new semiconductor materials. One such material, gallium nitride (GaN), is a compound semiconductor broadly used in the electronics industry for manufacturing LEDs, high frequency transistors, and other optoelectronic devices. Apart from its utility in electronic components, GaN possesses several properties conducive to ionizing radiation detection in harsh environments. To determine the utility of GaN as a radiation detector, vertical Schottky diodes were fabricated on commercially available, freestanding n-type GaN. Characterization of the electrical properties of the detectors was assessed by performing capacitance-voltage, current-voltage, temperature dependent current-voltage, and depth-resolved cathodoluminescence spectroscopy experiments on the devices. Alpha-particles from an Am-241 source were used to evaluate the charge collection efficiency and resolution of the detectors. Experiments were also performed to measure the neutron response of the detectors when using a Li-6:F conversion layer, as well as intrinsic neutron detection via neutron capture in N-14. Finally, neutron damage studies were carried out to assess the maximum fluence the detectors are capable of withstanding. Work will be presented detailing the fabrication of detectors from bulk GaN material, as well as experimental assessments of GaN's utility as a radiation detector.

Log: 302. **EFFECT OF IRON-BEARING ENVIRONMENT ON THE ALTERATION OF SON68 AND ISG GLASSES.** Lemesle, T.(1); Neeway, J.J.(2); Ryan, J.V.(2); Qafoku, N.P.(2); McCloy, J.S.(3); Wall, N.A.(1). (1) Washington State University, Chemistry Department, Pullman, WA, USA. (2) Pacific Northwest National Laboratory, Energy and Environment Directorate, Richland, WA, USA. (3) Washington State University, School of Mechanical and Materials Engineering, Pullman, WA, USA.

High level waste vitrification uses borosilicate glass matrix kept in iron alloy canisters for permanent storage in long-term repositories. This work aims at understanding the alteration of two simulated high-level waste glasses: SON 68 (non-radioactive composition of the French R7T7 glass used for fuel reprocessing) and International Simple Glass (ISG) (simplified SON68 with only 6 oxides) in an iron-bearing environment. These studies were focused on the effects of hematite (Fe<sub>2</sub>O<sub>3</sub>), a steel corrosion product, on glass alteration in four different solutions (18.2 MΩ water, NaCl 0.01 M, CaCl<sub>2</sub> 0.01 M and MgCl<sub>2</sub> 0.01 M) at 90 °C. The behavior of hematite in these solutions in absence of any glass was also tested. Solution concentrations and pH were measured and post-reaction solids were analyzed using SEM and XRD. For most systems, calculated normalized mass loss (NL) of each element and dissolution rates showed that the presence of hematite led to little or no change in the NL values or rate compared to a system without hematite. However, SON68 alteration in 0.01 M MgCl<sub>2</sub> in the presence of hematite demonstrated a slower increase of the pH and Mg concentration decrease, and a lower boron NL, indicating a lower alteration and precipitation kinetic. Furthermore, observation of higher pH and absence of soluble iron for samples

containing hematite can be due to carbonation phenomena. This work was supported by DOE/NEUP, under Project 23-3361.

Log: 303. **EFFECT OF SOLUTION COMPOSITION ON THE ALTERATION OF THE SON68 AND ISG GLASSES.** Lemesle, T.(1); Neeway, J.J.(2); Ryan, J.V.(2); Qafoku, N.P.(2); McCloy, J.S.(3); Wall, N.A.(1). (1) Washington State University, Chemistry Department, Pullman, WA, USA. (2) Pacific Northwest National Laboratory, Energy and Environment Directorate, Richland, WA, USA. (3) Washington State University, School of Mechanical and Materials Engineering, Pullman, WA, USA.

Borosilicate glass matrices are currently used for high-level radioactive waste immobilization in several countries. The high-level waste glasses, poured in canisters before storage in a long-term geologic repository, may come in contact with groundwaters within the repository. The aim of this work is to understand the alteration of two simulated high-level waste glasses (ISG and SON68) in the presence of salt-bearing aqueous solutions: NaCl 0.01 M and 5.00 M, MgCl<sub>2</sub> 0.01 M, CaCl<sub>2</sub> 0.01 M, and in distilled de-ionized water (18.2 MΩ). Both glass coupons and powders were used to achieve a Surface/Volume ratio of 40 cm<sup>-1</sup> or 60 cm<sup>-1</sup> (surface area measured by BET). Solution pH and cation concentrations were measured and solids analyzed by SEM and XRD. Normalized mass loss, alteration thickness and dissolution rates showed higher glass alteration in MgCl<sub>2</sub> solution. The average dissolution rates stabilize between 5E-4 and 5E-3 g/m<sup>2</sup>/d after 200 days of alteration at 90°C. Data obtained for systems in NaCl solutions revealed a lower normalized loss in 5 M NaCl compared to 0.01 M, which is interpreted as the accumulation of Na<sup>+</sup> at the negatively charged surface of the glass creating a dense layer that limits the release of elements in solution. This work was supported by DOE/NEUP, under Project 23-3361.

Log: 304. **EFFECT OF THE SURFACE/VOLUME RATIO ON THE ALTERATION OF SON68 AND ISG GLASSES.** Lemesle, T.(1); Neeway, J.J.(2); Ryan, J.V.(2); Qafoku, N.P.(2); McCloy, J.S.(3); Wall, N.A.(1). (1) Washington State University, Chemistry Department, Pullman, WA, USA. (2) Pacific Northwest National Laboratory, Energy and Environment Directorate, Richland, WA, USA. (3) Washington State University, School of Mechanical and Materials Engineering, Pullman, WA, USA.

The nuclear industry produces high level wastes that are managed by immobilization in borosilicate glass matrices confined in metal canisters for long-term storage in geologic repositories. This work aims to understand the alteration of the International Simple Glass (ISG) (6 oxide simplified glass composition based on SON68) and SON68 compositions (French borosilicate glass used for high level waste conditioning), at different glass-surface-to-solution-volume ratios. The experiments were performed using both glass monoliths and powders, at 90°C for 9 months, in four aqueous solutions – distilled de-ionized water (18.2 MΩ), NaCl 0.01 M, CaCl<sub>2</sub> 0.01 M, and MgCl<sub>2</sub> 0.01 M – in the presence or absence of hematite powder (a potential corrosion product of metal canisters) at glass/solution S/V ratio of 40 and 60 cm<sup>-1</sup>. Solution pH and analyte concentrations were measured, and the glass surfaces (monoliths and powder) were studied. Data showed that there is no effect of the S/V ratio on the pH values but the normalized mass loss is higher for 40 cm<sup>-1</sup> than for 60 cm<sup>-1</sup> experiments. The dissolution rates were evaluated to be between 5E-4 and 5E-3 g/m<sup>2</sup>/d. This work was supported by DOE/NEUP, under Project 23-3361.

Log: 305. **USING NOVEL TECHNOLOGY FOR SAMPLE SIZE REDUCTION IN PLUTONIUM METAL TRACE IMPURITY ANALYSIS.** Montoya, D; Borrego, A; Castro, A; Xu, N. Los Alamos National Laboratory.

The current Inductively Coupled Plasma – Optical Emission Spectroscopy (ICP-OES) instrumentation protocol for multi-elemental analysis in plutonium metal requires approximately 10 mL volume of sample. Our current facility is scheduled to be decommissioned by the year 2019. The new facility has a requirement that restricts the amount of radioactive materials used for analysis and requires a reduction in waste generation. In order to meet the stringent material limit requirements and still provide reliable data, new technology needs to be explored and new instrument method needs to be developed. In this study, an ICAP 6500 ICP-OES from Thermo Scientific was coupled with the ESI micro-FAST sample introduction system for sample size reduction. The integration of the two systems allowed for a sample size reduction of 88% for trace impurity analysis with improved detection limits. This allows the studies in Pu sustainment and nuclear forensics to be conducted continuously in the new facility while meeting the stringent radiological operations. LA-UR-14-29290



Log: 306. **DEVELOPMENT OF MICROCALORIMETER DETECTORS WITH EMBEDDED RADIOISOTOPES FOR DECAY ENERGY SPECTROSCOPY.** Mark P. Croce (1), Evelyn M. Bond (1), Andrew S. Hoover (1), Gerd J. Kunde (1), Veronika Mocko (1), Michael Rabin (1), Nina R. Weisse-Bernstein (1), Laura E. Wolfsberg (1), Douglas A. Bennett (2), James Hays-Wehle (2), Dan R. Schmidt (2), Joel N. Ullom (2). (1) Los Alamos National Laboratory. (2) National Institute of Standards and Technology.

Microcalorimeters with embedded radioisotopes are a promising new category of sensor for a variety of applications. Superconducting transition-edge sensor microcalorimeters can be combined with an absorber that contains embedded radioisotopes to perform decay energy spectroscopy with ultra-high energy resolution. For alpha-decaying isotopes such as Pu-239, Pu-240, and Am-241, this is a measurement of the total nuclear reaction energy ( $Q$ ) and the spectra consist of sharp, narrow peaks. In contrast, for beta decaying isotopes such as Pu-241, the spectrum is a low-energy continuum ending at  $E=Q$ . A central challenge is the encapsulation of the radioactive materials of interest inside the absorbing component in a way that ensures complete capture of the relevant decay products and eliminates spectral artifacts. By controlling the structure of the absorber matrix and radioactive materials, we are developing absorbers that enable high-resolution measurement of alpha- and beta-decaying isotopes for two specific applications: isotopic analysis of trace actinide samples, and neutrino mass measurement by decay energy spectroscopy of Ho-163. Analysis of trace actinide samples requires measurement of both alpha- and beta-decaying isotopes, with a detector dynamic range that spans keV to MeV energies. Measurement of Ho-163 requires detectors optimized for high resolution and high speed at the keV energy scale. We will discuss different techniques for radioactive material deposition and encapsulation along with sensor development and testing.

Log: 307. **MEASUREMENT OF CS DIFFUSION COEFFICIENT IN GRAPHITE USING ICP-MS AND NEUTRON ACTIVATION ANALYSIS.** Carter, L.M.(1); Brockman, J.D.(2); Robertson, J.D.(1) (2); Loyalka, S.K.(1). (1) University of Missouri. (2) University of Missouri Research Reactor.

High Temperature Gas-Cooled Reactors (HTGRs) are designed to operate at temperatures exceeding 1100 K and use helium as a primary coolant gas. These reactors are designed to use TRISO fuel particles which are approximately 1 mm in diameter and are coated with layers of graphite and silicon carbide that act as barriers to release of fission products. Measurements of the diffusion coefficients of fission products in graphitic materials used in the TRISO fuel and other core components are needed for modeling fission product release and transport during HTGR operation. In this work, we present a novel method to measure the diffusion coefficient of Cs in graphite. Spheres of graphite were uniformly loaded with Cs metal and analyzed using neutron activation analysis to determine the initial Cs mass loading. The Cs released from the graphite sphere was transported directly to an inductively coupled plasma mass spectrometer (ICP-MS) for real time release measurements using a carbon particle helium jet. Diffusion coefficients for Cs in IG-110 graphite in the 1100-1300 K temperature range were obtained by fitting the measured fractional release data with solutions to the diffusion equation. The observed values are in good agreement with previous measurements.

Log: 308. **High Precision Measurements of Stable Element and Actinide Isotope Ratios Employing Quadrupole (Q) and Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS).** James Sommers (1), Crystal Poole(2), Jeffrey Berg (3), Theresa Giglio (4), Gilles Youinou (5) and Jeffrey Giglio (6). (1-6) Idaho National Laboratory.

Neutron cross sections characterize the way neutrons interact with matter and are essential to most nuclear engineering projects undertaken. Current disciplines requiring low uncertainty neutron cross section information include: fission reactor design; nuclear fuel cycle; and nuclear safety and safeguards, among others. While there has been progress made into the theoretical predictability of measurement cross section models, the experimental determination of cross sections is vital to validation and improvement of the model data. This presentation will focus on the high precision measurements of stable elements, uranium and transuranic isotopes that have been irradiated in the Idaho National Laboratory's Advance Test Reactor by Q-ICP-MS and MC-ICP-MS. The dissolution and subsequent measurement of high purity, isotopic samples ranging from Ru through Cm-248 will be discussed. The optimization of the Q/MC-ICP-MS as well as a discussion of important instrumental parameters such as mass bias correction, hydride correction and electron multiplier cross calibration will be discussed. Analytical results for isotope ratios measured, including error determinations utilizing Guide to Uncertainty Measurement (GUM) software, will be presented. Lastly, future measurement plans as well as areas of improvement and further measurement plans will be discussed.

**Log: 309. AUTOMATIC IDENTIFICATION AND QUANTIFICATION OF RADIONUCLIDES IN GAMMA SPECTRA USING CONVEX OPTIMIZATION.**

Dayman, K. (1); Biegalski, S. (1) University of Texas at Austin.

Gamma spectrometry is a multi-elemental tool for nondestructively determining the composition of a sample from a single measurement; however, spectra taken of complex samples often present significant challenges to the analyst due to the large number of photopeaks and other spectral features typically observed. These features include interference, where the contribution from multiple radionuclides to a single observed peak must be resolved, sum peaks, and single- and double-escape peaks. We have developed a new algorithm and analysis method to pseudo-automatically identify and quantify the nuclides present in a sample using mathematical optimization. Our analysis features a peak-search to identify possible source nuclides taking energy calibration errors into account, applies heuristic criteria to identify possible sum and escape peaks, and then populates decay chains of radionuclides. These decay chains are fed into the optimization framework along with response data taken from an easily edited database we assembled and solved using convex optimization techniques. We have tested this methodology on several synthetic datasets as well as gamma spectra collected of medium- and long-lived fission products.

**Log: 310. EVALUATION OF INDEPENDENT AND CUMULATIVE FISSION PRODUCT YIELDS WITH GAMMA SPECTROMETRY.**

Dayman, K. (1); Biegalski, S. (1); Haas, D. (2); Prinke, A. (2); Stave, S. (2). (1) University of Texas at Austin. (2) Pacific Northwest National Laboratory.

Fission product yield data are critical to many applications; however, there are deficiencies existing reference data, most notably significant disagreement between datasets, inappropriate reported uncertainties that fail to capture the limitations of current knowledge, and a lack of existing measurements of independent yields. Using a Bayesian methodology combining gamma spectrometry, fission product modeling, convex optimization, and Monte Carlo sampling, we have made new measurements of independent and cumulative yields for a range of fission products. Fission products were produced using a 14 MeV generator at Pacific Northwest National Laboratory, as well as a pulsed neutron source at the Nevada National Security Site, to irradiate <sup>235</sup>U foils. Gamma spectra were collected for several days following the end of irradiation, and the data were fed into the Bayesian analysis. In this paper, we present the impetus for the measurement campaign, the methodology and validation exercises used to measurement independent and cumulative fission yields, and the results of measurements made to date.

Log: 311. **GAMMA AND BETA RADIOLYSIS OF NITRIC ACID: RADIOLYTIC NITRITE PRODUCTION.** Ibtihel Denden (1) ; Jean-Luc Roujou (2) ; Vincent Dauvois (2) ; Jean-Philippe Renault (1) ; Gerard Baldacchino (1). (1) CEA, DSM, IRAMIS, F-91191 Gif-sur-Yvette, France. (2) CEA, DEN, DPC, F-91191 Gif-sur-Yvette, France.

Radiolysis of nitric acid has been widely studied partly because of the use of HNO<sub>3</sub> as a medium in the processing of nuclear fuels. Nitrite ion is one of the major radiolytic species of HNO<sub>3</sub>. A particular interest is addressed to this ion owing to its implication in the solution redox properties. In this framework, the purpose of this work is to focus on parameters influencing the production of NO<sub>2</sub><sup>-</sup> ions. Therefore, a series of nitric acid solutions were exposed to  $\gamma$  and electron radiations in order to reveal the impact of HNO<sub>3</sub> concentration, dose rate and presence of platinoids matrix (Pd, Ru, and Rh). It was shown that G(NO<sub>2</sub><sup>-</sup>) increases from 10<sup>-10</sup> to 10<sup>-8</sup> mol/J with increasing HNO<sub>3</sub> concentration. However, in the presence of palladium platinoids, G(NO<sub>2</sub><sup>-</sup>) drastically increases with regard to irradiated solutions that don't contain Pd. Radiolytic yields are ranging between 10<sup>-8</sup> and 10<sup>-7</sup> mol/J depending on HNO<sub>3</sub> concentration. Observable G(NO<sub>2</sub><sup>-</sup>) were simulated by using CHEMSIMUL code, a differential-equation system solver applied to chemical reaction system. As an important preliminary result, these simulations exhibit a change of NO<sub>2</sub><sup>-</sup> concentrations with increasing dose rates. The exposition of HNO<sub>3</sub> solutions to  $\gamma$  rays and electron beams in the presence of various fission products matrix (Sr, Tc, Cs, Co, Ru...) will provide independent influences in chemical competition. These studies are still in progress.

Log: 312. **A PROTOTYPE DIGITAL COINCIDENCE SYSTEM FOR THE MEASUREMENT OF GAMMA EMISSIONS FROM ALPHA-GAMMA EMITTERS IN HIGH RATE GAMMA BACKGROUND ENVIRONMENT.** SCIVIER P.C. AWE PLC.

A PROTOTYPE ALPHA-GAMMA COINCIDENCE DETECTION SYSTEM HAS BEEN DEMONSTRATED TO OVERCOME MEASUREMENT ISSUES AND INTERFERENCES THAT ARISE FROM UNSEPARATED MIXED ALPHA-GAMMA SOURCES AND HIGH RATE GAMMA BACKGROUND SOURCES, SUCH AS THOSE GENERATED FROM FISSION REACTIONS. 239PU AND 241AM ALPHA-GAMMA EMITTERS WERE CHOSEN TO DEMONSTRATE THE SENSITIVITY IMPROVEMENTS THAT CAN BE ACHIEVED OVER CONVENTIONAL GAMMA SPECTROMETRY METHODS IN THESE CHALLENGING ENVIRONMENTS. THE SYSTEM UTILISES A HPGE GAMMA DETECTOR & PASSIVE IMPLANTED PLANAR SILICON DETECTOR, BOTH TIME SYNCHRONISED USING DIGITAL MULTI CHANNEL ANALYSERS. IT HAS BEEN SHOWN THAT GREATER THAN 99 PERCENT REDUCTION OF A HIGH GAMMA BACKGROUND CAN BEEN ACHIEVED, WITHOUT THE PRESENCE OF A LEAD SHIELD. THE USE OF THIS COINCIDENCE TECHNIQUE INCREASES CONFIDENCE LEVELS FOR ALPHA EMITTING NUCLIDE IDENTIFICATION, QUANTIFICATION AND MINIMUM DETECTABLE ACTIVITY LEVELS, WHEN COMPARED TO CONVENTIONAL HPGE GAMMA SPECTROMETRY.

Log: 313. **A Blueprint for Radioanalytical Reference Materials – 2014.** Inn, K.G.W.(1); Nour, S.(2); LaRosa, J.(2); Mann, J.(2); LaMont, S.P.(3); Jerome, S.(4); Essex, R.(3); Johnson, C.M.(5); Leggitt, J.(6); Morrison, J.(7). (1) K&E Innovations. (2) NIST. (3) USDOE. (4) NPL. (5) US Army. (6) FBI. (7) DHS.

Reference materials are fundamental tools used by radiochemistry measurements laboratories to establish and evaluate analytical methods, test measurement capabilities, compare analytical results over time and between programs, and establish legal confidence in measurement results. Over the past decades low-level environmental reference materials have been developed mostly for environmental monitoring and geosciences. However, more recently radionuclide reference materials have been developed for other communities such as occupational safety, ocean studies, food safety, nuclear power decommissioning, and environmental restoration. Currently, new national and international priorities has resulted in demands for improved, broader, and new reference materials for urban remediation and recovery, radiobioassay, emergency consequence management, and nuclear forensics, treaty monitoring, non-proliferation and environmental safeguards, and other future needs. With the application of new generation mass spectrometry to complement traditional radioactivity counting the new reference materials will need to be certified on both the radioactivity and mass basis. It is important to keep in mind that developing good reference materials is a complex process that requires careful metrology and may take a considerable amount of time with a commiserate expense. In this current financial climate, there are limited resources for the material producers, so every effort should be made to carefully balance production costs with the need to serve as many customers as reasonable. This requires communication, coordination and funding among all programs and countries to move the reference material effort, and ultimately the quality of analytical measurements, into the future.

Log: 314. **ACCEPTANCE CRITERIA FOR RADIONUCLIDE MEASUREMENT TRACEABILITY PROFICIENCY TESTING MATERIALS.** Inn, K.G.W. K&E Innovations.

It insufficient for laboratories to demonstrate traceability through the possession of SRMs, traceable reference materials, or instrument calibrations because of the potential for improper use. Independent testing of measurement traceability is the only mean by which it is truly realized. An issue that remains is the criteria by which traceability is evaluated to be successfully demonstrated. While the ANSI N42.22 acceptance criteria (and similar criteria) to demonstrate measurement traceability, i.e., difference less than three times the uncertainty, is becoming more widely used, there is also a need to evaluate the acceptability of the test materials used during the tests as a check for the testing laboratory. It is proposed that a difference between the mean result from tested laboratories and the reference value greater than  $1.21 s(\text{lab})$  [ $\alpha = 0.05$ ] or  $1.9 s(\text{lab})$  [ $\alpha = 0.01$ ] would indicate a potential bias and the testing laboratory should investigate the situation for an erroneous supply of test materials.

Log: 315. **HOW OLD IS IT? - <sup>241</sup>PU/AMNUCLEAR FORENSIC CHRONOLOGY REFERENCE MATERIALS.** Fitzgerald, R. (1); Inn, K.G.W.(2); Horgan, C.(3). (1) NIST. (2) K&E Innovations. (3) Worcester Polytechnic Institute.

Nuclear forensics attempts to characterize nuclear materials, either retrieved from post-detonation debris or secured before an incident. Through this analysis and the determination of such qualities as material age, impurity content, and ratios of radioisotopes, one seeks to determine the origin of the material in question. From there, it may be possible to identify those involved and take steps to ensure national security and prevent malicious use or attribute the perpetrators. However, to determine information such as the amount of time that has passed since the material's last purification, a reliable standard is required. <sup>241</sup>Pu is almost always present in uranium- and plutonium-based nuclear weapons, which pose the greatest threat to our security. The in-growth of <sup>241</sup>Am due to the decay of <sup>241</sup>Pu provides an excellent chronometer of the material. It is apparent that a well characterized <sup>241</sup>Pu/<sup>241</sup>Am standard is needed to validate measurement capability, as a basis for between-laboratory comparability, and as material for verifying laboratory performance. This effort verifies the certification of a 34 year old <sup>241</sup>Pu Standard Reference Material (SRM4340) through alpha-gamma anticoincidence counting, and confirms the validity of the original separation date to two weeks.

Log: 316. **ARE THEY THE SAME? RETROFITTING 1G SAMPLE DATA TO CERTIFIED 10G SAMPLE CHARACTERIZATION.** Filliben, J.(1), Inn, K.G.W.(2), Simon Waldren, S.(3). (1) NIST. (2) K&E Innovations. (3) AWE.

NIST Ocean Sediment SRM 4357 was certified for anthropogenic radionuclides with sample size of > 10 g. The distribution of the anthropogenic radionuclides measurements were best described with a Weibull function. However, measurement by mass spectrometry offers high sensitivity and the use of smaller samples. Plutonium measurements at Aldermaston used a 1-g sample size and resulted in a highly skewed measurement distribution with larger than expected number of results outside of the upper end of the certified tolerance range for 10-g samples. Were the results from 1-g samples equivalent to those from 10-g samples used to certify the material? Three batches of results were examined: 1) the original set of 1-g Aldermaston plutonium results submitted for the certification of the material; 2) the original set of 10-g plutonium results submitted for the certification of the material; and 3) a new set of 1-g Aldermaston QC plutonium results. By batching 10 1-g sample Aldermaston results into the equivalence of a 10-g sample result, then conducting distributional fitting, it was found: 1) the original Aldermaston 1-g results were equivalent to the new QC 1-g results; and 2) the 10 1-g batched Aldermaston results were equivalent to the 10-g measurement results used to originally certify the plutonium in SRM 4357. It was concluded that the Aldermaston remained in control between their two sets of measurements, their results were consistent with the SRM 4357 certified plutonium values, and using smaller sample size than the recommended magnifies the sample heterogeneity for uncertainty assessment.

Log: 317. **ANALYSIS OF URANIUM AND THORIUM IN SOILS AND SEDIMENTS - HOW COME WE SOMETIMES GET THE WRONG RESULT?**. Inn, K.G.W. K&E Innovations.

NIST has certified a number of soils and sediments for uranium and thorium isotopic content over the past three decades. The certification process involves international intercomparison of experienced laboratories. In general, the laboratories add their tracers, use acid leaching, acid dissolution or fusion techniques to dissolve the sample, a combination of ion exchange and ion chromatography separations, and then alpha spectrometry for measurement (occasionally, a laboratory would use mass spectrometry for measurement). At times we have found no difference among laboratory results, and at other times there are differences based on sample dissolution technique used. In one case, Rocky Flats Soil - I (SRM 4353), there was significant difference (15%) even between acid dissolution (HNO<sub>3</sub>, HCl, HF) and KF fusion. It is suspected that there are minor (<0.03% by sample mass) Zr-related mineral fractions containing relatively high concentrations of U and Th that resists HF treatment, unless the process is carried out over many days. This issue is confirmed in related work to be environmentally widespread and requires vigilance by the analyst.

Log: 318. **USDOE - CIAE INTERNATIONAL NUCLEAR FORENSICS COOPERATION ON AGE-DATING URANIUM**. Schorzman, K.C. (1); Kinman, W.S. (2); Steiner, R.E. (2); Cardon, A.M.R. (2); Williams, R.W. (1); Yonggang, Z. (3). (1) Lawrence Livermore National Laboratory. (2) Los Alamos National Laboratory. (3) China Institute of Atomic Energy.

In 2014 the China Institute of Atomic Energy (CIAE) and the USDOE/NNSA Office of Defense Nuclear Nonproliferation (NA-21) International Nuclear Forensics Cooperation Program (INFC) began a cooperative study in the area of uranium age dating measurements. Lawrence Livermore National Laboratory (LLNL) and Los Alamos National Laboratory (LANL), representing INFC, and CIAE agreed to share their procedures for the radiochemical purification and mass spectrometry determination of <sup>230</sup>Th and <sup>234</sup>U in bulk uranium samples and to analyze uranium standard reference materials U010 and U850. The aim of this study is to improve DOE/NNSA and CIAE laboratory capabilities for measuring the age of bulk uranium materials using the <sup>230</sup>Th/<sup>234</sup>U radiochronometer. Precise and accurate results for age-dating depend on the calibration of the spike materials (e.g., <sup>229</sup>Th and <sup>233</sup>U) used for the isotope dilution mass spectrometry (IDMS) analyses. To facilitate inter-comparison of results, samples of a <sup>230</sup>Th standard reference material were distributed to each laboratory for IDMS using their <sup>229</sup>Th spikes. The preliminary results from this cooperative study will be presented and the model-age results for U010 and U850 will be compared with previously published age-dates, and with the known production dates of these materials.

Log: 319. **THE SEPARATION OF AMERICIUM FROM CURIUM BY ION EXCHANGE CHROMATOGRAPHY.** Bond, E. M. (1); Moody, W. A.; Bredeweg, T. A. (1); Coleman, M. E. (1); Slemmons, A. K. (1); Vieira, D. J. (1). (1) Los Alamos National Laboratory. The development of a separation of americium from curium will be described. The research was conducted in order to support the measurement of the neutron capture isomer ratio for Am-241. We explored three methods for the separation of americium from curium. First we explored the separation on cation exchange resin after oxidation of the americium from the trivalent to the pentavalent state. Next, we studied the separations using cation exchange chromatography in acidic alpha-hydroxyisobutyric acid solutions. Finally, we examined the separation of americium from curium using methanol/nitric acid solutions. This last method was the method chosen for the separation of the irradiated Am-241 target materials; the results of the separations of the tracer levels of curium from mg quantities of Am will be discussed. LA-UR-14-29377

Log: 320. **SPATIALLY RESOLVED ULTRA-TRACE ANALYSIS OF URANIUM AND PLUTONIUM.** Hamann, L. (1); Dubchak, S. (2); Walther, C. (1). (1) Institut für Radioökologie und Strahlenschutz, Leibniz Universität Hannover, Germany. (2) State Ecological Academy of Post-Graduate Education and Management, Kyiv, Ukraine.

In today's environment a significant amount of radionuclides released by testing of nuclear weapons, reprocessing plants, nuclear reactors or accidents, is present in the form of radioactive particles of various size, composition and specific activity. As the release rates of these particles are generally very low, analysis techniques with highest sensitivities are needed. Previous studies show that a pure trace analytical detection for example of plutonium is not sufficient for all research questions. Due to the complex chemical bonding behavior detailed speciation studies are essential in order to obtain a deep insight of its behavior in the environment. The time-of-flight secondary-ion mass-spectrometry (TOF-SIMS) allows the spatially resolved analysis of radionuclides in the sub-micron scale with simultaneous detection of all major and minor elements of the sample. With a TOF-SIMS solid surfaces are analyzed by sputtering the surface with a focused primary ion beam. Targeted electron bombardment can prevent charging the surface. This approach allows analyzing of non-conductive environmental samples without any chemical treatment, which could destroy the structure of the radioactive particles. In this work samples from Chernobyl and Fukushima are analyzed for uranium and plutonium containing particles. To find these particles, the samples were preselected by gamma spectroscopic analyses and solid state nuclear track detection and then analyzed by a TOF-SIMS.



Log: 321. **DETERMINATION OF PLUTONIUM AND URANIUM IN ENVIRONMENTAL SAMPLES FROM FUKUSHIMA.** Schneider, S. (1); Christl, M. (3); Steinhäuser G. (2); Walther, C. (1). (1) Leibniz Universität Hannover, Institut für Radioökologie und Strahlenschutz, Germany. (2) Department of Environmental and Radiological Health Sciences, Colorado State University, United States of America. (3) Laboratory of Ion Beam Physics, ETH-Zürich, Switzerland.

During the Fukushima accident significant amounts of radioactive nuclides were released into the atmosphere, including small amounts of actinides. Previous investigations showed a strong localization of plutonium, which indicates a particulate release of this element from the NPP. As plutonium is of a high radiological relevance if incorporated, further investigations on vegetation, litter and soil samples taken at eight different sampling sites in the vicinity of the damaged reactor were conducted. Accelerator mass spectrometry is a powerful tool to determine even lowest concentrations. For these measurements the samples were first leached and then uranium and plutonium were chemically separated using extraction chromatography. Here we use the U-236/U-238 and the Pu-240/Pu-239 isotopic ratios as indicators to distinguish between global fallout and releases from a nuclear reactor.

Log: 322. **RADIOLAB PROJECT: RADON INDOOR MEASUREMENTS A WAY TO EDUCATE TO NUCLEAR ISSUE.** Groppi, F. (1); Bonardi, M.L. (1); Manenti, S. (1); Gini, L. (1); Bazzocchi, A. (1,2). (1) Università degli Studi di Milano and INFN, Physics Department – LASA Laboratory. (2) Itis Mattei.

Since 2005 in Italy two programs were promoted from Ministry of Education and University and National Institute of Nuclear Physics (INFN) for scientific dissemination in the field of environmental radioactivity, devoted to High School students that are involved in particular in indoor radon measurements. The idea of this project started from the consideration that the subjective perception (sensation) of the risk doesn't correspond very often to the objective and real risk of human activity. In particular, this is true for the radioactivity theme that is often misleading because of the lack of accurate information. A way to approach this subject, to make the public more trusting of nuclear issues related also to energy and medical applications is to discuss about radioactivity and ionizing radiation. Starting from an experimental approach the students can receive basic but correct information on radioactivity subject. They are involved as a real research project participating to all the experimental phases, from detectors preparation, data acquisition, analysis and presentation to the public. The focus of the project is the "laboratory", intended as a method to "know" through the "know how", a new way to teach and to learn. The laboratory is not only a physical place, where the students are directly involved in experimental activities, but also a method to learn and to acquire skills in physics. The different aspects and some of the many results derive from this kind of project, carried forward for 10 years, will be presented.

Log: 323. **SPECIATION ANALYSIS OF PU AND U COMPLEXES IN SOLUTION USING MASS SPECTROMETRY AND LASER SPECTROSCOPY.** Walther, C. (1) University Hannover.

Many radionuclides have a very rich – some may call it complicated - solution chemistry. This is particularly true for some of the actinides. Under certain chemical conditions as many as four different oxidation states of Plutonium are present in solution simultaneously, each of them having unique properties. Consequently, quantification of the chemical species with regard to oxidation states, complexation, polymerization and also sorption onto or incorporation into solids needs to be investigated. Chemical species can vary over a wide size range, from small ions or molecules with sizes of less than 1 nm via small molecules with simple ligands to macromolecules, organometallic compounds, polymeric species, nano-particles and colloids ranging from 1 nm all the way to macroscopic mineral particles and other solid phases with sizes of more than 1µm. The species of different size ranges are related to each other, often in terms of chemical equilibria. One simple example is the equilibrium between two oxidation states determined by the redox potential. Measurements on Pu and U complexes with organic ligands in aqueous solution are conducted using Electrospray Mass Spectrometry (ESI-MS) for the stoichiometry of molecules, Time-Resolved Laser Spectroscopy (TRLFS) for chemical speciation of luminescent species, and Laser-Induced Breakdown Spectroscopy (LIBD) for quantification of aquatic colloids.

Log: 324. **Sequential Determination of Plutonium, Neptunium, and Americium in Environmental Samples.** Hanson, S. K.; Oldham, W. J.; Miller, J. L.; Pollington, A. D. Chemistry Division, Los Alamos National Laboratory.

A method has been developed for the sequential determination of plutonium, neptunium, and americium in environmental soil and sediment samples. The optimized procedure involves purification of plutonium and neptunium by anion exchange chromatography and measurement by ICP-MS using isotope dilution. The americium-containing fraction is collected, purified with both cation and anion exchange columns, and then measured by ICP-MS using an americium-243 isotope dilution standard. The ability to measure all three elements out of the same aliquot and the short time frame required for analysis are significant advantages of the method. The method has been used to determine neptunium, plutonium, and americium concentrations in several environmental matrix standard reference materials available from NIST, including SRM 4357 (Radioactivity Standard).

Log: 325. **Kläui Ligand Thin Films for Rapid Plutonium Analysis by Alpha Spectrometry.** Hanson, S. K.; Oldham, W. J. Chemistry Division, Los Alamos National Laboratory.

A new method for binding plutonium for analysis by alpha spectrometry has been developed. Thin films of Kläui-type tripodal oxygen donor ligands were prepared by spin-casting solutions onto glass substrates. Three different ligands were evaluated for plutonium binding and the best results were obtained using an ethyl-substituted Kläui complex, which bound 80-88% of the dissolved Pu under equilibrium conditions. The thin films are simple and inexpensive to prepare and exhibit excellent alpha spectral resolution, having line widths of ~33 keV. The method has been successfully applied to analyze for plutonium in both an archived nuclear debris sample and a certified environmental soil sample. The results obtained from the soil analysis are in good agreement with the certified values, demonstrating the effectiveness of the method for rapid plutonium analysis.

Log: 326. **RECENT INTERNATIONAL ASSISTANCE IN NUCLEAR FORENSICS.** Smith, D.K.(1); Bull, T.(1); Tsvetkov, T.(1); Liu, D.(1); Mrabit, K.(1). (1) Division of Nuclear Security, International Atomic Energy Agency.

Nuclear forensics is accepted as the scientific analysis of nuclear or radioactive material as well as associated evidence, which can assist in law enforcement investigations and nuclear security vulnerability assessments. Data characteristics, or signatures, imparted into materials throughout the nuclear energy spectrum to include the fuel cycle may be exploited to provide information on the origin and history as well as the diversion of material found out of regulatory control. Over the past decade, the International Atomic Energy Agency (IAEA), working with its Member States and international partners, has developed a program of nuclear forensics assistance for the prevention of and response to a nuclear security event. More than 85 Member States have participated in IAEA nuclear forensics training offered regionally and internationally. In addition, coordinated nuclear forensic research has focused on instrumentation and methodologies to include gamma spectroscopy and mass spectrometry, laboratory procedures for analysis of bulk materials as well as microparticles, interpretative methods for identification of nuclear fuel cycle process history, modeling of plutonium and uranium isotope signatures from spent nuclear fuels, and application to samples obtained from throughout the nuclear fuel cycle. Published technical guidance on the conduct of a nuclear forensics examination includes the forthcoming 'Nuclear Forensics in Support of Investigations' to be published as part of the IAEA Nuclear Security Series. In 2014, the first ever IAEA International Conference on Advances in Nuclear Forensics served as a successful forum for States to exchange recent experience on common approaches to and effective implementation of nuclear forensics.

Log: 327. **COMPREHENSIVE CHEMICAL ANALYSIS OF AMERICIUM-241 OXIDE IN SUPPORT OF LARGE-SCALE PRODUCTION OPERATIONS.** Castro, A.; Borrego, A.P.; Montoya, D.P.; Spencer, K.J.; Stanley, F.E.; Xu, N.; Tandon, L.; Schappert, M.F.; Martinez, A.; Actinide Analytical Chemistry, Los Alamos National Laboratory.

Domestic US supplies of Am-241 have dwindled over the past 25 years. The greatest demand for this isotope is fueled by the oil and natural gas production industries, as AmO<sub>2</sub>/Be neutron sources are ideal for well-logging purposes. High isotopic purity Am-241 can be recovered from plutonium which has been separated from irradiated fuel, and then allowed to age prior to separation of ingrowth Am-241. Both weapons-grade Pu and reactor-grade Pu isotopic mixtures include a fraction of Pu-241, with reactor-grade having a greater fraction of the heavier Pu isotopes. The beta decay of Pu-241 (half life of 14.4 years) to Am-241 provides a path to high isotopic purity material in Pu that has been cleanly separated from irradiated fuel. This decay path provides a close tie between Am-241 production campaigns and Pu reprocessing operations in all historical DOE efforts to produce AmO<sub>2</sub>. The Actinide Analytical Chemistry Group at LANL is developing chemical analysis methods in support of the current DOE AmO<sub>2</sub> production campaign. Various methodologies are being established to determine Am-241 chemical and isotopic purity and trace element contamination, including Pu content and isotopic composition. A centralized dissolution/separation/aliquoting scheme has been developed for subsequent analysis by an array of techniques and instrumentation, including thermal ionization mass spectrometry (TIMS), inductively coupled plasma -atomic emission spectroscopy (ICP-AES) and -mass spectrometry (ICP-MS). The results for the analysis of AmO<sub>2</sub> pilot samples are presented here.

Log: 328. **MORPHOLOGICAL IMAGES AND CHEMICAL SPECIATION OF UF<sub>6</sub> HYDROLYSIS PRODUCTS.** Wilkerson, M. P.; Wagner, G. L.; Kinkead, S. A.; Paffett, M. T.; Rector, K. D.; Scott, B. L.; Tamasi, A. L.

Characterization of chemical speciation and morphologic properties of products from hydrolysis of uranium hexafluoride (UF<sub>6</sub>) is important for understanding dispersion and potential health consequences from inhalation of released material. Uranium hexafluoride is a gray crystalline solid with a high vapor pressure under standard temperature and pressure. Due to its volatility, it is essential for isotope separation, and as such, may be a 'fingerprint' for breakout from reactor- or fuel-grade to weapons-grade uranium. A number of studies report complex chemical reactivity between UF<sub>6</sub> and water vapor, and suggest that relative humidity and temperature play a role in the chemical and physical composition of the hydrolysis products and the evolution of these products over time. Additional work suggests that deposition surfaces may have a role in the morphology of the hydrolysis products formed. Our limited understanding of the complexity of these systems, however, precludes our ability to develop viable and quantitative signatures from these products. An understanding of these properties will lead to a more effective understanding of the detectable signatures from UF<sub>6</sub> hydrolysis. We have developed a method for producing uranium oxy-fluoride material, and we have exposed these materials under controlled ambient conditions of temperature and relative humidity. We will discuss morphology images, vibrational spectroscopic measurement, and powder X-ray diffraction analyses from these products. LA-UR-14-29330

Log: 329. **SIMULTANEOUS PRODUCTION OF MULTIPLE CARRIER-FREE RADIOTRACERS USING 50 MEV PROTONS.** Finn, E.C.; Wittman, R.; Greenwood, L.R.; Woods, V.; Bowen, J.M.; Seiner, B.N. Pacific Northwest National Laboratory.

Waste waters commonly contain mixtures of many different toxic elements, including nickel, copper, and lead. Gamma-emitting radiotracers of these elements can be utilized in applications such as environmental analysis, toxicological studies, and more. However, gamma-emitting radiotracers of nickel and lead radioisotopes are not commercially available at this time, in part due to the short half-lives of the radionuclides <sup>57</sup>Ni and <sup>203</sup>Pb. Copper radiotracer <sup>67</sup>Cu may be purchased commercially, but the production of a copper radiotracer was compatible with lead and nickel isotopes production and is thus considered here. Gallium radiotracer <sup>67</sup>Ga is a usable byproduct of copper production. In this work, carrier-free radiotracers of nickel, lead, copper, and gallium, which are readily detected by their favorable gamma decay properties, were produced simultaneously by irradiation of a cobalt-thallium-zinc foil stack by 50 MeV protons at the University of California, Davis cyclotron. The attenuation of the beam energy passing through each foil was utilized to simultaneously optimize production of each of the desired radiotracers. Future work will include improvement of radiochemical separations to rapidly isolate the short-lived radiotracers from by-products and unspent target material. Simultaneous production of multiple radiotracers in a cyclotron is a unique application.

Log: 330. **DEUTERON ACTIVATION ANALYSIS OF A PALLADIUM IMPURITY IN PLATINUM.** Finn, E.C.; Wittman, R.; Greenwood, L.R.; Friese, J.I. Pacific Northwest National Laboratory.

Deuteron irradiation of a Pu-240 source electrodeposited on a platinum disc resulted in production of Am-240 as well as several activation products of the platinum disc. Gamma spectrometry measurements on the platinum disc after recovery of the Pu and Am identified Au-194 through Au-199, as well as the silver isotopes Ag-106m, -110m, -111, and -112. The production of Au isotopes conformed to the activity predicted for deuteron activation of platinum based on both published cross-sections and EMPIRE II models. The production of silver isotopes was not predicted prior to bombardment, but was traced to the presence of a palladium impurity in the platinum disc. Based upon published cross-sections for deuteron activation of palladium and the mass of the platinum disc, the weight percent of palladium in the platinum disc was calculated.

Log: 331. **SURROGATE POST-DETONATION URBAN DEBRIS STANDARD REFERENCE MATERIAL.** Mann, J.L. (1); Tyra, M.A. (1); Molloy, J.L. (1); Buscaglia, J. (2); Pfeuffer, K. (2); Dettman, J. (2); Leggitt, J. (2); Kelly, T.D. (3); McClory, J.W. (3); and Jerome, S. (4). (1) NIST. (2) FBI. (3) AFIT. (4) NPL.

The discipline of nuclear forensics was developed because of an increasing concern that nuclear or radiological material will be used in a terrorist attack. Nuclear forensics is the thorough collection, analysis, and evaluation of radiological and nuclear material in pre-detonation and post-detonation scenarios resulting from a nuclear detonation. Conclusions drawn from this collected data coupled with law enforcement and intelligence information may support attribution – the identification of those individuals responsible for planned and actual attacks. Reference materials (RMs) establish the traceability, accuracy, and precision of nuclear forensic measurements and provide the foundation to stand up to legal scrutiny. Current RM development efforts, in collaboration with partner organizations (DHS DNDO NTNFC, FBI, DoD, DOE National Laboratories, and international partners), focus on producing certified and/or working RMs for nuclear forensic analysis that include, for example, RMs for mass spectrometric calibration, isotopic tracer RMs for isotope dilution mass spectrometry (IDMS), and radiochronometry RMs. NIST, in concert with partner labs (AFIT (DoD), FBI (DOJ) and NPL) and support from the FBI, have developed a Surrogate Post-Detonation Urban Debris Standard Reference Material (SRM). This test material has been developed to mimic the “rubble” of a city after an Improvised Nuclear Device (IND) detonation, which is capable of producing fresh fission products. We are in the process of assessing the homogeneity of the minor and trace elements in the material by energy dispersive X-ray fluorescence spectrometry and neutron activation analysis, respectively. The next stages will be certifying these elements, as well as the U isotope composition.

Log: 332. **NEW APPLICATIONS OF KNiFC-PAN RESIN FOR BROAD SCALE MONITORING OF RADIOCESIUM FOLLOWING THE FUKUSHIMA DAI-ICHI NUCLEAR DISASTER.** Breier, C.F. (1); Pike, S.M. (1); Sebesta, F. (2); Breier, J.A. (1); Singh, H. (1); Buessler, K.O. (1). (1) Woods Hole Oceanographic Institution. (2) Czech Technical University.

Since the 2011 tsunami and Fukushima nuclear power plant (FNPP) disaster off the coast of Japan, there has been a strong public interest and need to continue to measure and monitor cesium radioisotopes in the ocean. Concern for monitoring of radiocesium along the U.S. West Coast prompted testing for new applications of a resin that will allow for large scale monitoring studies. Tests of KNiFC-PAN, potassium-nickel hexacyanoferrate (II) (KNiFC) bound into modified polyacrylonitrile (PAN) have shown that this resin is capable of absorbing radiocesium with up to 98% recovery. To collect Cs, a filtered 20 L seawater sample is passed through 5 ml of KNiFC absorber that is then dried and counted on a germanium well detector. Since 2012 this method has been used to measure Cs over several cruises along the Japan coastline. This method has also been employed in a crowd-funded citizen scientist campaign (OurRadioactiveOcean.org) to measure Cs along the west coast of the U.S. and on a transect from Alaska to California. To monitor Cs over a larger spatial scale, a method that can achieve faster collection of samples in-situ is required. An in-situ sampler is being modified for this purpose. This sampler will be deployed on an autonomous vehicle (JetYak) for broader scale sampling. Additionally, devices containing KNiFC-PAN that can be worn or attached to personal watercraft are being developed with the goal of increasing public participation in the monitoring of radiocesium in the ocean.

Log: 333. **Determination of long-lived radionuclides in decommissioning waste of nuclear reactors.** HOU Xiaolin (1), (2) (1) Technical University of Denmark, Center for Nuclear Technologies (2) Xi'an AMS Center, Institute of Earth Environment, CAS, China.

For characterization of various wastes from decommissioning of nuclear reactors, it is required to determine all radionuclides, especially long-lived radionuclides for evaluation of the radioactivity inventory in various materials and decision making for management of the produced waste. In the waste depository view, the most important radionuclides of reactor waste are those long-lived and high mobile radionuclides. Among them the most important are  $^{36}\text{Cl}$ ,  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{237}\text{Np}$  and plutonium isotopes. Besides the radiometric methods, mass spectrometric techniques are attractive method for the measurement of long-lived radionuclides. In the recent years, ICP-MS and AMS are intensively used for the measurement of long-lived radionuclides. Because of their low concentration in most of samples and their beta or alpha decay without measurable gamma rays, chemical separation of these radionuclides from the sample matrix, as well as from other interferences are the major challenges for their accurate determination. We have established a series of analytical method for the determination of these radionuclides in various types of waste from the decommissioning of nuclear reactors. This presentation will summarize these works by focusing on both measurement techniques and chemical separation procedures.

Log: 334. **MEASUREMENTS OF TRACE ACTINIDE SIGNATURES ASSOCIATED WITH THE LOCATION OF A FORMER MANHATTAN PROJECT ERA PLUTONIUM FACILITY IN LOS ALAMOS, NEW MEXICO.** Oldham, W.J.; Matteson, B.S.; Hanson, S.K.; Miller, J.L. Los Alamos National Laboratory.

Trace level plutonium mobilized in the environment due to natural weathering processes is efficiently adsorbed onto the surface of rusty metal objects. Radiochemical analysis of the surface oxide coating found on discarded metal objects such as bottle caps or common nails can provide a sensitive indicator of nearby nuclear activities. In this project, rusty bottle caps of contemporary origin were collected from sites around northern New Mexico and were analyzed for trace plutonium contamination by isotope dilution inductively-coupled plasma mass spectrometry (ICP-MS). Bottle caps collected from remote locations show plutonium isotopic ratios consistent with mixtures of global and regional fallout from atmospheric nuclear testing ( $\text{Pu-240/Pu-239} = 0.168 \pm 0.012$ ). In contrast bottle caps collected near the site of the former Manhattan Project Era plutonium facility (Technical Area 1; D building) reveal a unique isotopic signature indicative of the earliest plutonium manufactured as part of the U.S. war effort ( $\text{Pu-240/Pu-239} = 0.0012 \text{ to } 0.0070$ ). These results suggest that analysis of the surface oxide coating from rusty metal objects can provide a sensitive detection strategy to enhance the effectiveness of environmental monitoring programs.

Log: 335. **DISTRIBUTION OF NEPTUNIUM AND PLUTONIUM IN NEW MEXICO LICHEN SAMPLES (USNEA ARIZONICA) CONTAMINATED BY ATMOSPHERIC FALLOUT.** Oldham, W.J. (1); Hanson, S.K. (1); Lavelle, K.B. (2); Miller, J.L. (1). (1) Los Alamos National Laboratory. (2) University of Cincinnati.

*Usnea* is a widely distributed, yellowish-green fruticose genus of lichen. Occurring within montane regions of Arizona and New Mexico, the species *Usnea arizonica* (western bushy beard) typically grows on ponderosa and pinion pine trees. As a rule, lichens obtain essential nutrients directly from the atmosphere and have evolved highly efficient mechanisms to bioconcentrate trace elements within their tissues. This characteristic has been used for many years in both Europe and North America to monitor the distribution of atmospheric pollutants. In this paper, the concentration and isotopic composition of Np-237, Pu-239, and Pu-240 were measured using isotope dilution inductively-coupled plasma mass spectrometry (ICP-MS) in samples of *Usnea arizonica* retrieved from remote locations in New Mexico, USA. Consideration of the  $\text{Pu-240/Pu-239}$  and  $\text{Np-237/Pu-239}$  ratios indicates likely source terms from both global and regional fallout resulting from atmospheric nuclear testing. Regional fallout from the Trinity test and atmospheric testing at the Nevada Test Site (circa 1951 – 1962) significantly reduces the observed  $\text{Pu-240/Pu-239}$  and  $\text{Np-237/Pu-239}$  ratios. The fact that actinide contamination continues to be detected in recent lichen collections suggests continuous re-suspension of fallout radionuclides even 50 years after ratification of the Limited Test Ban Treaty.



Log: 336. **ISOTOPIC PURITY EVALUATION OF THE ZR-93 ENRICHED SAMPLE.** Fujii, T.(1); Hori, J.(1); Du, M.(2); Fukutani, S.(1); Takamiya, K(1); Yamana, H(1); Kiyanagi, Y(3). (1) Kyoto Univ. (2) ORNL. (3) Hokkaido Univ.

A project to determine the neutron-capture cross section of long lived fission products and minor actinides has been started by using a beam-line at Japan Proton Accelerator Research Complex (J-PARC). One of the target nuclides is zirconium-93 (Zr-93), whose half-life is 1.5E6 years. The sample of 0.47 g enriched Zr-93, which was fabricated and imported from Oak Ridge National Laboratory (ORNL), USA, is used in the experiment. A small amount of the same zirconium material was imported together in order to perform qualitative and quantitative analyses at Research Reactor Institute, Kyoto University (KURRI), Japan. Radioactive impurities were analyzed by gamma spectrometry. The isotopic composition of Zr-m (m: 90, 91, 92, 93, 94, and 96) was precisely determined by multi-collector thermal ionization mass spectrometry with < 0.1% of 2 sigma uncertainty. To increase the reliability of nuclear data obtained, the validity of our purity evaluation is discussed.

Log: 337. **TOWARD THE AQUEOUS CHEMISTRY OF COPERNICIUM UTILIZING HOMOLOGUE SEPARATIONS.** Mudder, P. R. (1,2); Nitsche, H. (1,2). (1) University of California Berkeley. (2) Lawrence Berkeley National Laboratory.

Over the past twenty years, the periodic table has been greatly expanded due to new element discoveries, with livermorium, element 116, being the heaviest confirmed element. With the exception of gas-phase studies of elements bohrium (107), hassium (108), copernicium (112), and flerovium (114), nothing is known about the aqueous chemistry of elements 107 to 116. Chemical studies of these elements are limited due to their short half-lives and extremely low production rates, requiring the capability for fast atom-at-a-time studies. We have begun to study the aqueous chemistry of Cn through its homologues Cd and Hg. A bench top liquid-liquid separation scheme was developed with phosphine oxide ligands in toluene selectively extracting the Cd and Hg in different aqueous conditions. We are in the process of developing a new liquid-liquid separation device that will accommodate separations on a 100 millisecond timescale. We plan to test this device with longer lived homologue isotopes first in batch experiments and later with cyclotron-generated shorter lived isotopes.

Log: 338. **ROUND-ROBIN  $^{230}\text{Th}$ - $^{234}\text{U}$  AGE DATING OF BULK URANIUM FOR NUCLEAR FORENSICS.** Gaffney, A.M. (1); Hubert, A. (3); Kinman, W.S. (2); Magara, M. (4); Okubo, A. (4); Pointurier, F. (3); Schorzman, K.C. (1); Steiner, R.E. (2); Williams, R.W. (1). (1) Lawrence Livermore National Laboratory. (2) Los Alamos National Laboratory. (3) Commissariat à l'Énergie Atomique et aux Énergies Alternatives. (4) Japan Atomic Energy Agency.

The model age of nuclear material is an important signature that can be used in a nuclear forensic investigation to constrain the production history and origin of the material. We have completed a tri-lateral round robin measurement exchange to compare analytical methods for  $^{230}\text{Th}$ - $^{234}\text{U}$  age dating of bulk uranium materials. The  $^{230}\text{Th}$ - $^{234}\text{U}$  model ages are determined assuming a model history wherein (i) the  $^{230}\text{Th}$  decay product was purified completely from the bulk U material at the time that the material was produced, and (ii) the bulk U material remained a closed system since the time of its production. If the U material was incompletely purified from Th at the time of production, then the model date will precede the actual production date, and thus the model age will be a maximum age for the material. In this investigation,  $^{230}\text{Th}$ - $^{234}\text{U}$  model ages of NBL CRM U050 were determined by four laboratories. All measurements utilized an isotope dilution mass spectrometry method (IDMS). Uranium IDMS analyses were performed on TIMS and MC-ICP-MS instruments, and Th IDMS measurements were performed on MC-ICP-MS and HR-ICP-MS instruments. The model ages determined range from 3-May-1956 to 3-August-1957, and all age determinations made by the four labs agree within the respective analytical uncertainties. Although some individual age determinations have uncertainty estimates that overlap the paper age, all of the model ages are older than the paper age of October, 1957. This may reflect trace  $^{230}\text{Th}$  impurities remaining in the bulk material after production. LLNL-ABS-665283.

Log: 339. **DESORPTION OF CESIUM FROM MINERALS: THE EFFECT OF MINERAL COMPETITION.** Durrant, C.B. (1,2); Begg, J.D. (1); Zhao, P. (1); Zavarin, M. (1); Unlu, K. (2); Kersting, A.B. (1). (1) Glenn T. Seaborg Institute, Lawrence Livermore National Laboratory. (2) Pennsylvania State University.

Understanding adsorption and desorption processes is essential for predicting the mobility of radionuclides in the environment. Typically, adsorption/desorption reactions are studied in single mineral systems. Here we investigate adsorption and desorption of cesium in mono and bi-mineral experiments in an attempt to move towards understanding radionuclide desorption in complex mineral environments. A Cs isotherm experiment was performed with illite, a non-expanding micaceous clay mineral, and Cs-137 doped over the concentration range  $10\text{E-}3\text{ M}$  to  $10\text{E-}10\text{ M}$ . After the initial adsorption period, batch desorption experiments were performed with the isotherm samples using  $10\text{E-}3\text{ M}$  of non-radioactive cesium as the background electrolyte and using Cs-137 as a tracer for desorption. In separate experiments, Cs was sorbed onto illite, montmorillonite, and kaolinite for at least 7 days. Aliquots of the mineral suspensions were then placed inside 5 mL dialysis bags with a pore size cut-off size of 3.5-5 kD. The dialysis bags were then placed inside cells containing a 175 mL mineral suspension of illite with a known concentration of cesium outside of the dialysis bag. The desorption of cesium initially adsorbed to either the illite, montmorillonite, kaolinite placed inside the dialysis bag was monitored by measuring the amount of Cs-137 that diffused into the bulk suspension outside the dialysis bag. After optimizing these desorption experiments with the non-redox active cesium, we will attempt to do a similar study with plutonium. Ultimately we aim to use our data to improve adsorption/desorption parameters for environmental transport models and evaluate the impact of desorption rates on radionuclide transport. This research was performed under the Nuclear Forensics Graduate Fellowship Program, which is sponsored by the U.S. Department of Homeland Security, Domestic Nuclear Detection Office and the U.S. Department of Defense, Defense Threat Reduction Agency. This material is based upon work supported by the U.S. Department of Homeland Security under Grant Award Number, 2012-DN-130-NF0001-02 and the U. S. DOE Office of Biological & Environmental Sciences, Subsurface Biogeochemistry Research Program. LLNL-ABS-664666. The views and conclusions contained in this document are those of the authors and should not be interpreted as representing the official policies, either expressed or implied, of the U.S. Department of Homeland Security.

Log: 340. **DEVELOPMENT OF AN AUTOMATED CHEMICAL SEPARATION SYSTEM FOR FORENSIC SAMPLE ANALYSIS.** Durrant, C.B. (1,2); Despotopulos, J.D. (1); Henderson, R.A. (1); Moody, K.J. (1); Unlu, K. (2); Shaughnessy, D.A. (1). (1) Lawrence Livermore National Laboratory. (2) Pennsylvania State University.

Nuclear forensic analysis primarily seeks to determine the origin and composition samples. Being able to do this rapidly has proven challenging. One of the primary concerns of analyzing in situ forensic samples is the ability to determine the composition of the sample, particularly the fuel type and corresponding fission products. Through radiochemical separations this information could be ascertained and significantly increase the rapidity with which the question of origin and composition could be answered. Here an automated chemical separation system that is being developed for the analysis of nuclear forensic samples is presented. This system attempts to address the problem of being able to obtain radiochemical data from in situ forensic samples quickly and efficiently. This separation system was initially tested with a mixture of uranium, tin, and lead isotopes to show the effectiveness and efficiency of the separation system. This research was performed under the Nuclear Forensics Graduate Fellowship Program, which is sponsored by the U.S. Department of Homeland Security, Domestic Nuclear Detection Office and the U.S. Department of Defense, Defense Threat Reduction Agency. This material is based upon work supported by the U.S. Department of Homeland Security under Grant Award Number, 2012-DN-130-NF0001-02. The views and conclusions contained in this document are those of the authors and should not be interpreted as representing the official policies, either expressed or implied, of the U.S. Department of Homeland Security. LLNL-ABS-664635.

Log: 341. **RADIATION MEASUREMENT SYSTEMS AND EXPERIENCES IN JAPAN AFTER THE FUKUSHIMA ACCIDENT.** Bronson, F. L.; Canberra.

In the approximately 4 years since the March 2011 earthquake and subsequent Tsunami damaged the three Dai-Ichi nuclear power plants, the AREVA and Canberra team have deployed a variety of different radiation measurement machines in Japan. These measurement projects or instruments include: • Dose-rate mapping of the close-in areas on the damaged reactor site; • Spectroscopic nuclide-specific on-line measurements of the first water processing unit on the site; • General purpose food measurement units, both high precision HPGe systems and low cost NaI systems; • In-vivo measurement systems, both fixed and mobile whole body counting units, that were used on at least 500,000 people; • In-vivo measurements with special high-sensitivity system for babies and children; • Automatic systems for high throughput assay of bags of rice from Fukushima Prefecture; • High sensitivity automatic system to assay boxes of special semi-dry persimmons; • System to assay soil on a conveyor belt and sort the output according to level of radioactivity; • System to assay a truck loaded with 1 cubic meter sacks of soil and vegetation; • On-line water measurement system for SrY90 at levels suitable for release to the environment; • Mobile system to prove that rolls of grass for animal feed meet the regulatory requirements. This document briefly describes the purpose of each measurement project, describes the instruments and assay method that used, and discusses some of the operational experiences from each of these projects.

**Log: 342. COMPARISON OF MASS SPECTROMETRIC METHODS (TE, MTE & CONVENTIONAL) FOR URANIUM ISOTOPE-AMOUNT RATIO MEASUREMENTS.**

K. J. Mathew (1) and A. Hasozbek (1\*) (1) New Brunswick Laboratory, Department of Energy (\*) present address: Dokuz Eylul University, Izmir-Turkey.

New Brunswick Laboratory (NBL) of the United States Department of Energy is the certifying authority for uranium and plutonium assay and isotopic standards used in various stages of the nuclear fuel cycle. NBL provide certified reference materials (CRMs) traceable to the international system of units (S. I.) that can be used for calibration of analytical equipment used by nuclear analytical facilities. The safeguards measurement evaluation program administered by NBL provides an external validation of the quality control practices of nuclear analytical facilities. NBL also contribute to development of analytical techniques routinely used by nuclear facilities for assay and isotope ratio measurements. Recent contributions include: i) the modified total evaporation technique for isotope amount ratio measurements using TRITON, ii) the direct total evaporation technique for MAT261 with Spectromat ® hardware and software upgrades, and iii) the development of the uranium isotope dilution mass spectrometry technique using NBL CRMs, other than <sup>233</sup>U, as spikes. We present a comparison of the isotope amount ratio measurement techniques routinely used for characterization of CRMs using thermal ionization mass spectrometer (TIMS) instruments. Three different uranium isotope amount ratio measurement techniques - total evaporation (TE), modified total evaporation (MTE), and Conventional analysis techniques - are compared. The advantages of each method, the fractionation profiles resulting from the measurement techniques, and factors affecting systematic components of bias in the isotope amount ratio measurements are presented. Dominant components of uncertainties as it relates to these measurement techniques and the estimation of GUM compliant uncertainties are discussed.

**Log: 343. ALPHA RADIOLYSIS OF PLUTONIUM SOLUTIONS IN NITRIC ACID.**

Balasubrahmanyam Srinivasan, Nancy Hui, and Alma Stiffin. New Brunswick Laboratory, U. S. Department of Energy.

A number of glass vessels containing plutonium solutions are stored in the New Brunswick Laboratory (NBL) vault complex designated as a HC-3 facility. The solutions represent a potential source of revenue if converted to Certified Reference Materials (CRMs) to support measurements in nuclear safeguards, environmental and forensic samples analyses. Since NBL is in the process of downgrading its operation to radiological status, these solutions are to be "shipped" to another HC-3 facility at the Argonne site for "temporary" storage and retrieval later for conversion to CRMs. The integrity of the solutions must be preserved during shipping and while in storage at the new facility. An option being considered is to ship and store the solutions in tightly stoppered glass containers to prevent mass loss. Under these conditions, the pressure build up due to gaseous products from alpha radiolysis must be evaluated as a part of safety and hazards analyses; the expected gaseous products are hydrogen, oxygen, nitrogen, and nitrogen oxides. Results of this evaluation will be presented for the (isotopically) different plutonium solutions in

storage, all in nitric acid, by taking into account their composition, energy release in radioactive decay, G-values, and free volume. Note that the inherent economic value present in these plutonium solutions in storage can be easily realized through conversion to CRMs needed by the US and international customers, if they are preserved properly. Similar evaluation was done for U-233 solution (CRM 111-A) in sealed glass ampoules and these results will be included in the presentation.

Log: 344. **AGE-DATING OF SINGLE PARTICLE OF URANIUM STANDARD MATERIAL FOR NUCLEAR FORENSICS AND SAFEGUARDS.** Okubo, A., Esaka, F., Magara, M., Shinohara, N.; JAEA.

To elucidate the final purification date of uranium material namely age-dating is important subject on nuclear forensic analysis. Age-dating of uranium using  $^{234}\text{U}$ - $^{230}\text{Th}$  pair nuclide was established in the field of geochemical science, and has been applied for nuclear forensics. Recently, safeguards analysis also demands age-dating of uranium materials. Because of relatively young age of uranium materials (less than ca. 60 years), difference of the abundance between uranium and thorium reaches more than nine order of magnitude. In order to measure such a small amount of  $^{230}\text{Th}$  by ICP-MS, anion exchange column separation is commonly conducted to eliminate uranium tailing effect. However, column separation step could raise procedural blank of  $^{230}\text{Th}$ . In contrast, TIMS can separately ionize uranium and thorium in filament heating step using the difference of optical ionization temperature of these elements. We therefor developed analytical method for age-dating of single particle uranium material by TIMS. Single particles of uranium standard material (CRM U-500) were sampled separately by micro-sampling devices inside a scanning electron microscope. The collected particle samples were decomposed by nitric acid, and spiked with  $^{233}\text{U}$  and  $^{229}\text{Th}$ . In order to reduce thorium blank, rhenium filaments were thoroughly baked out before use. Uranium isotopic ratios were previously measured using secondary electron mode (SEM). After reducing uranium ionization, the thorium isotopic ratios were measured. The accuracy of measurements were 1-3% RSD for  $^{234}\text{U}/^{233}\text{U}$  and 7-8% RSD for  $^{230}\text{Th}/^{229}\text{Th}$ , respectively. The determined age in this work was reasonably agreed with expected age given in literature.

Log: 345. **SEASONAL VARIATION OF ATMOSPHERIC DEPOSITION OF BE-7 AND H-3 IN KOREAN AMBIENT AIR AND RAIN.** Yoon, Y.Y.; Koh, D.C.; Lee, K.Y; Cho, S.Y. Korea Institute of Geoscience and Mineral Resources.

Be-7 is a cosmogenic radionuclide produced in the stratosphere and troposphere as a result of cosmic ray spallation of nitrogen and oxygen. It has a relatively short half-life ( $T_{1/2} = 53.3$  days) and emits a gamma-ray of 477.6 keV. Shortly after production, these isotopes become attached to aerosols and are transported through the atmosphere and ultimately, via precipitation, to the Earth's surface, where they enter soils, marine sediments and ground water. In this study, we observed the seasonal variation of the Be-7 activities in air by a continuous sampling with a high volume air dust sampler. And also, Be-7 and H-3 activities in precipitation were determined. The monthly activity level of Be-7 in air was ranged from 1.94 to 47.2 Bq/m<sup>3</sup>. Be-7 in the precipitation was separated using cation exchange resin and the activity level was ranged from 0.29 to 0.99 Bq/L. H-3 was analyzed using electrolytic enrichment method and activity ranged from 0.50 to 2.22 Bq/L

Log: 346. **Compound specific radiocarbon analysis from indoor air samples via accelerator mass spectrometry.** Kretschmer, W.; Schindler, M.; Scharf, A.; Tschekalinskij, A. Physics Department, University of Erlangen, Erwin-Rommel-Str. 1, Erlangen 91058, Germany.

Many volatile organic environmental compounds are potentially dangerous due to their allergic or carcinogen impact on humans. For the establishment of effective countermeasures for lowering their concentration in houses, sources have to be known. Our investigation is focused on aldehyde compounds since their indoor concentration is often above the official guidelines and since they originate from biogenic or anthropogenic sources. Both types of sources can be distinguished by their different <sup>14</sup>C content which can be measured via accelerator mass spectrometry (AMS). For the collection and separation of these gaseous substances they have to be converted into liquid or solid phase by derivatization. This leads to the incorporation of up to six additional carbon atoms into the derivatized sample and hence to a reduced <sup>14</sup>C content. To reduce the number of additional carbon atoms and to optimize efficiency and duration of the procedure, different derivatization compounds and methods have been tested with acet- and formaldehyde of known <sup>14</sup>C content. The Erlangen AMS facility, based on an EN tandem accelerator and a hybrid sputter ion source for solid and gaseous samples, is well suited for the measurement of isotope ratios  $^{14}\text{C}/^{12}\text{C} \approx 10^{-12} - 10^{-15}$ . The <sup>14</sup>C concentration of the calibration samples and from indoor air samples in apartments, beer taverns and schools have been determined by AMS, the corresponding results are discussed with regard to potential sources of aldehydes.

Log: 347. **ATTENUATION, EFFICIENCY, AND ESCAPE LINES.** Coldwell,RL,(Coldwell consulting) (1) Lasche,GP, (Snakedance Scietific(TM), LLC) (2).

The authors have developed and tested a method to determine the efficiency of a detector by approximating it as essentially one minus the attenuation in the detector. The attenuation is a known function of the energy of a gamma ray, the distance that it travels in the detector, the density of the detector and the effective charge  $Z$  of the detector. Most gamma rays enter the detector after attenuation outside the detector so that the total attenuation is a combination of that in the detector and that outside the detector. Energetic gamma rays react with the nuclei to produce electron positron pairs inside the detector that subsequently produce 511 KeV gamma ray pairs. Some of these gamma rays escape the detector resulting in single and double escape lines. The escaping gamma rays are effectively attenuated only in the detector. This means that the sizes of the escape lines enable the detector attenuation to be determined separately from the overall attenuation. Assumptions and detail will be given and the results tested by fits to various spectra.

Log: 348. **EXPRESS NONDESTRUCTIVE GEOCHEMICAL ASSAY BY INSTRUMENTAL PHOTON ACTIVATION ANALYSIS WITH SHORT-TIME IRRADIATION USING THE MT-25 MICROTRON.** Krausová, I.(1); Mizera, J.(1,2); Řanda, Z.(1); Chvátíl, D.(1); Horák, Z.(1); Krist, P.(1). (1) Nuclear Physics Institute, Academy of Sciences of the Czech Republic. (2) Institute of Rock Structure and Mechanics, Academy of Sciences of the Czech Republic.

The MT 25 microtron is a radio frequency cyclic accelerator of electrons that can be used as an effective source of secondary high energy photon radiation (bremsstrahlung) in the instrumental photon activation analysis (IPAA). Recent installation of an automated pneumatic system for rapid sample transport between the beam position and detector in the short time, online irradiation mode, provide substantial extension of the analytical range of IPAA particularly in express, nondestructive assay of major elements for geochemical analysis via photonuclear, mainly ( $\gamma$ , n) and ( $\gamma$ , p) reactions with short-lived products. Procedures for the express basic geochemical analysis have been developed and optimized with a representative set of geochemical reference materials: CRPG-CNRS BEN (Basalt), GS-N (Granite), and UB-N (Serpentine), and USGS QLO-1 (Quartz Latite) and AGV-2 (Andesite). Optimizing beam energy and irradiation-decay-counting times allowed reliable IPAA determination of the elements Si, Fe, K, Mg, Al, Zr, and Ba in all selected USGS and CRPG-CNRS geochemical reference materials, and was applied also in analysis of real geological samples. The work has been supported by the project 13-27885S of the Czech Science Foundation.



Log: 349. **PRECISE AND ACCURATE U ISOTOPE ANALYSIS BY MODIFIED TOTAL EVAPORATION (MTE) USING 1E13 OHM RESISTOR AMPLIFIERS ON A THERMO SCIENTIFIC™ TRITON PLUS™ MC-TIMS.** Trinquier, A.(1); Komander, P.(1); Bouman, C.(1); Schwieters, J.(1); Douthitt, C.(2). (1) Thermo Fisher Scientific, Bremen, Germany. (2) Thermo Scientific, USA.

This study reports the first use of 1E13 Ohm amplifiers with a TIMS for measurement of extreme U isotopic ratios. The performance of the MTE protocol developed by NBL, IAEA-SGAS, ITE, and IRMM was assessed using the IRMM-187 certified U standard (Richter et al. 2011 JAAS 26, 550). Sub-microgram sample loads were run in multiple static multicollection mode, combining 1E11 Ohm amplifiers for the major U ion beams with 1E13 Ohm amplifiers on <sup>234</sup>U, <sup>236</sup>U ion beams and the 233.7, 234.4, 235.7, and 236.4 half masses. Each measurement comprised three ion beam cup-settings, allowing analysis of U isotopes and a per mass cycle subtraction of half-masses, for optimized correction of peak tailing effects from the major isotopes. The best internal precision was 0.1 ‰ on <sup>234</sup>U/<sup>238</sup>U and 0.3 ‰ on <sup>236</sup>U/<sup>238</sup>U (2RSE). External reproducibilities for static multicollection analysis of the NBS-U010 certified standard using an SEM and a 1E13 Ohm amplifier were compared. Microgram sample loads were run at constant current intensity. Because of the high stability of the 1E13 Ohm current amplifiers and the fact that there is no ion counter/Faraday cup cross calibration uncertainty, external reproducibility for ion beams ≥ 62 kcps is twice as good with a 1E13 Ohm amplifier (0.2%) compared to an SEM (0.4%). Faraday cups using high gain amplifiers provide accurate and precise analysis of uranium isotopic ratios including small abundance isotopes, down to ion beams in the 10 kcps range.

Log: 350. **LEAD-BISMUTH – FISSION PRODUCTS INTERACTION.** Espegren, F.; Retegan; T. Chalmers University of Technology: Nuclear Chemistry and Industrial Materials Recycling.

The concept “Defense in Depth” concerns several levels of protection, like including several successive barriers for preventing the release of radioactive material to the environment. As it is well known, the fuel matrix itself, and the integrity of the fuel rod and the boundary of the primary coolant system are the first physical barriers against the potential release of radionuclides. Gen IV reactors designs have already addressed the barrier represented by the primary coolant system. In this study, the focus has been on the lead-bismuth eutectic (LBE) interaction with fission products. The fission products investigated in this project were I<sub>2</sub>, Cs and Ru in contact with LBE at different temperatures (200°C, 300°C and 400°C) with ingress of air. The behaviour of coolant-fission product interaction is important for knowing how the formed compounds potentially could behave in a reactor, e.g. if they are immobile and remain close to the breach of the cladding tube or if they follow the flow of the coolant and agglomerate in another part of the reactor vessel. The results of the investigation have shown that some interaction happens between the LBE and all the fission products at all temperatures in air, where at the two lower temperatures Bi was predominating at interacting with the fission products, compared to Pb, thus being immobilized in LBE at an early stage.

Log: 351. **EVALUATING TREE RINGS AS ARCHIVES OF ENVIRONMENTAL URANIUM CONTAMINATION.** McHugh, K.C.(1); Widom, E.(1); Spitz, H.B.(2); Glover, S.E.(2). (1) Miami University. (2) University of Cincinnati.

Concentrations and isotopic ratios of metals in tree rings have recently been employed as a tool to resolve temporal changes in anthropogenic inputs to the environment. However, this approach is compromised if metals translocate across annual growth rings, a process that is metal- and tree-species dependent. Few studies have explored the behavior of U in tree rings. The former Fernald Feed Materials Production Center, SW Ohio, provides an opportunity to compare the U record in tree rings to historical and sediment core records. This site produced U metal from natural ores and recycled nuclear materials from 1951-1989, with releases to the environment of 90 - 450 Mg U. Site remediation began in 1991 and continues today with groundwater treatment. Recent studies of lichen and tree bark point to the persistence of regional U contamination. For this study, fifteen samples containing four growth rings each from a sugar maple (*Acer saccharum*) core collected near site were analyzed by TIMS. Growth rings spanning 1945-2004 have low U concentrations (<3 ppb) but rings from the onset of the remediation phase (1989-1996) have up to 13 ppb U. Additionally, the core is characterized by natural  $^{235}\text{U}/^{238}\text{U}$  in pre-production and the earliest years of production (1945-1956), but depleted signatures post-1956. Likewise,  $^{234}\text{U}/^{238}\text{U}$  activity ratios of  $\sim 1.00$  (consistent with natural waters) are found prior to 1965, but are generally  $<1.00$  thereafter. These data are consistent with limited mobility of U in sugar maple and suggest that this species may be a robust archive of U contamination.

Log: 352. **CONFIRMATION OF CRM 125-A, A LIGHTLY - ENRICHED U RADIOCHRONOMETRIC CERTIFIED REFERENCE MATERIAL.** Tyra, M. A.(1), Mann, J. L.(1), LaRosa, J.(1), Nour, S.(1), Inn, K (1). (1) National Institute of Standards and Technology.

The National Institute of Standards and Technology (NIST) is performing confirmatory measurements on the lightly-enriched uranium Certified Reference Material CRM 125-A; New Brunswick Laboratory (NBL) recently re-characterized and re-certified this material to improve its utility as a U-234 - Th-230 nuclear forensic radiochronometric standard. We modified the prescribed experimental design and added additional chemical purification steps than was done for the NBL certification. Additionally, alpha spectrometric counting times were extended for each sample to improve the counting statistics of each measurement. As alpha-particle spectra deconvolution is a major contributor to uncertainty, care was taken to deconvolute each alpha-particle spectrum. We deconvoluted the U alpha-particle spectra with a modified DAMM (ORNL) code. We deconvoluted the Th alpha-particle spectra, however, manually—the Th-229 peak shape confounds our DAMM script due to the many decay pathways of Th-229 decay, U results are consistent with values reported in NBL's certificate.

Log: 353. **NEW OPTIONS TO IMPROVE TALSPEAK-BASED SEPARATIONS OF TRIVALENT ACTINIDES AND LANTHANIDES.** Kenneth L. Nash; Jenifer C. Braley(1); Derek M. Brigham(2); Travis S. Grimes(3); Colt R. Heathman(3); Aaron T. Johnson(3); Joseph L. Lapka; Mikael A. Nilsson(4); Cecile Marie (5). Washington State University, Department of Chemistry, P.O. Box 644630, Pullman, WA 99164-4630. Present addresses: 1. Colorado School of Mines, 2. Oak Ridge National Lab, 3. Idaho National Lab, 4. University of California at Irvine, (5) CEA, Marcoule, France. Several solvent extraction processes for the separation of trivalent fission product lanthanides from transplutonium actinides have been developed based on the enhanced actinide selectivity of polyaminopolycarboxylic acid complexing agents. Each of these processes is based on chemistry first proposed by Weaver and Kappelmann in the Trivalent Actinide-Lanthanide Separation by Phosphorus reagent Extraction from Aqueous Komplexes (TALSPEAK) process, which was developed at Oak Ridge National Laboratory in the 1960s. In the baseline process, Am/Nd separation factors near 100 are seen – Am separation from all other lanthanides is even more efficient. During the past 10 years research focusing on the chemistry of TALSPEAK has attempted to improve understanding of the complex solution chemistry that both enables and limits performance of TALSPEAK-based An/Ln separations. This increased knowledge of the solution chemistry has pointed the way toward improved TALSPEAK-based separations. In this presentation selected features of the fundamental chemistry of TALSPEAK will be discussed leading ultimately toward a description of new options and concepts designed to further improve prospects for large-scale application of TALPEAK chemistry in less conventional systems. Work supported at Washington State University by the U.S Department of Energy, Office of Nuclear Energy, Fuel Cycle Research and Development Program in several Nuclear Energy University Programs (NEUP) projects and within the Sigma Team for Minor Actinide Separations project.

Log: 354. **THE INVESTIGATION OF MODIFIED KALMAN FILTERS FOR DECONVOLUTION OF COMPLEX PEAK SHAPES IN THE MOSSBAUER SPECTROSCOPY OF ENVIRONMENTAL SAMPLES.** Abrecht, D.G. (1); Kukkadapu, R.V. (2); McDonald, B.S.(1); Schwantes, J.M.(1); Eiden, G.C.(1); Sweet, L.E.(1). (1) National Security Directorate, Pacific Northwest National Laboratory. (2) Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory.

The deconvolution and fitting of peaks is a critical component to the identification and characterization of environmental samples in Mossbauer spectroscopy. Unfortunately, because of the strong likelihood of peak overlap and typically low signal-to-noise ratio inherent to the technique, long count times are often required to obtain adequate statistics for performing good deconvolutions. In our previous work (Abrecht et al., Nucl. Instrum. Methods Phys. Res., Sect. A, 2014), we examined the use of software filters based on modifications of a Kalman filter to reduce signal-to-noise in the Mossbauer spectra of standard bcc iron foil, leading to reduced count times to obtain similar signal-to-noise ratios in the final spectra. In this work, we explore the use of this method for samples containing multiple environments for the target nucleus, with an emphasis on improving identification of overlapping peaks and reducing signal acquisition time for identification.

Log: 355. **ENHANCED TECHNIQUES FOR ULTRA-LOW LEVEL (PG AND FG) ACTINIDE ANALYSIS BY MC-ICP-MS.** Pollington, Anthony D.; Kinman, William S.; Hanson, Susan K. Nuclear and Radiochemistry, Los Alamos National Laboratory. Recent advances in instrumentation have led to the ability to measure high-precision isotope ratios at increasingly low analyte concentrations. Combining techniques for enhanced ionization with better counting of small ion beams, we routinely measure isotope ratios on 100's of pg uranium samples and  $\leq 10$  pg plutonium samples with relative standard deviations of 0.1% on major isotope ratios and 1% on minor ratios achievable. With slightly larger samples ( $\leq 1$  ng total U), these precisions can be as low as 0.01% and 0.1% respectively. The analytical setup is a Cetac Aridus II desolvating nebulizer interfaced with a ThermoScientific Neptune Plus equipped with a jet-type sample cone and x-type skimmer cone, leading to an increase in sensitivity on the order of 10x that of a standard spray chamber ( $\sim 1000$ V/ppm U). The Neptune Plus is equipped with 9 Faraday cups (FC) and 5 electron multipliers (EM), allowing for the simultaneous collection of all isotopes of either U or Pu with a combination of FC (e.g.,  $^{235}\text{U}$  and  $^{238}\text{U}$ ) and EM (e.g.,  $^{234}\text{U}$  and  $^{236}\text{U}$ ) with other configurations also available (e.g.,  $^{235}\text{U}$  and  $^{238}\text{U}$  can be measured on EM in small samples). As sample sizes get small, contributions from environmental blanks, as well as interfering species, become increasing concerns. In this study, we will present data on efforts to minimize the contribution of environmental U using scaled-down chemical procedures as well as the effect of polyatomic species on the precision and accuracy of actinide isotope measurements and what procedures can be applied to minimize interferences.

Log: 356. **Capabilities of an On-site Inspection.** Miley, H.S.(1); Haas, D.A.(1). (1) Pacific Northwest National Laboratory.

The authors will sketch the technical capabilities of a 10-person team operating a mobile lab and portable equipment against a nuclear anomaly that could be a contained underground nuclear explosion. Survey, including flight, carborne, and backpack, help locate an area for investigation, then in-situ survey and sample collections can find isotopic anomalies in concentration, location, and ratio. Where surface radionuclides are not evident, sub-surface noble gas (Xe and Ar) collection and mobile lab measurement can detect leakage from even well-contained nuclear tests. The authors will discuss the initial expectations vs realities of a 4-week field exercise.

Log: 357. **AN ATMOSPHERIC RELEASE OF IA-140 TO SIMULATE A SMALL-SCALE VENT FROM AN UNDERGROUND NUCLEAR EXPLOSION.** Keillor, M.E.(1); Milbrath B.D.(1); Arrigo, L.M.(1); Baciak, J.E.(2); Chipman, V.D.(3); Emer, D.F. (3); Detwiler, R.S.(1); Kernan, W.J.(1), Kirkham, R.R.(1); MacDougall, M. (1,4); Rishel, J.P. (1); Seifert, A.(1), Seifert, C.E.(1); Smart, J.E. (1). (1) Pacific Northwest National Laboratory. (2) University of Florida. (3) National Security Technologies (4) Oregon State University.

Researchers at Pacific Northwest National Laboratory have performed an experiment to simulate the near-field deposition pattern of radionuclides released in a small-scale vent from an underground nuclear test. High-purity lanthanum oxide powder was activated in the Washington State University research reactor to produce short-lived La-140. The particulate source material was injected into the atmosphere using a carbon dioxide gerb ("air cannon") in the Yucca Flats area of the Nevada National Security Site. The experiment successfully produced a narrow ground plume extending ~1.5 km downwind from the release point, with sufficient activity to compare and contrast several techniques of gamma radiation survey, and environmental sampling followed by gamma assay in a field laboratory. The techniques studied are relevant to methods planned for use by an On-Site Inspection (OSI) team under the verification regime of the Comprehensive Nuclear-Test-Ban Treaty. This work will present details and results of the experiment, including the source production, transport, release, sampling and survey, and will present lessons-learned that are relevant to the conduct of an OSI.

Log: 358. **MEASUREMENT BACKGROUND AND THE SEDIMENT AGE-DATING REACH OF SI-32.** Keillor, M.E.(1); Aalseth, C.E.(1); Arrigo, L.M.(1); Brandenberger, J.M.(1); Cloutier, J.M.(1); Eiden, G.C.(1); Fast, J.E.(1); Finch, Z.S.(1); Gill, G.A.(1); Hossbach, T.W.(1); Seiner, B.N.(1); Strandquist, S. (1). (1) Pacific Northwest National Laboratory.

Silicon-32 has been identified as an isotope that fills the sediment geochronology gap between Pb-210 at <150 years and C-14 at >1000 years. However, detector sensitivity and chemical isolation challenges of radiometric Si-32 for age dating have resulted in a fairly limited number of applications of this technique to real-world sediment studies. The significance of targeting the intermediate age bracket between 100 and 1000 years is to provide accurate geochronological reconstructions of paleoindicators that identify human and large-scale, climate-induced shifts in coastal areas. As an example, we are preparing new radiation detectors and kilogram-scale sample preparation techniques for a study of sediments recovered from Puget Sound, Washington, United States. Implications of detector background on the duration of required measurements and the age-dating reach are considered. The design of a low-background, gas-proportional beta counter for measurement of the daughter isotope P-32 is discussed; the detector background goal for these new detectors is ~10 counts per day, based on results of the sensitivity study. Current background and detection efficiency performance of the new beta detectors will be presented.

Log: 359. **METHODS TO IMPROVE XENON QUANTIFICATION.** Suarez, R. (1); Hayes, J.C. (1); Harper, W.W. (1); Humble, P (1); Ripplinger, M.D.; Stephenson, D.E. (1); Williams, R.M (1). (1) Pacific Northwest National Laboratory.

The precision of the stable xenon abundance in a gas sample is critical to accurately determine the activity concentration. Whole air gas processing systems, such as Swedish Unattended Noble gas Analyzer (SAUNA), that are part of the International Monitoring System (IMS) network use the stable xenon quantity as part of a beta-gamma measurement to determine the radioactivity present in a gas sample. These types of systems typically use a plastic scintillating beta-cell as part of the radiation measurement. One of the challenges in the stable xenon quantification measurement is accounting for gas mixing and xenon uptake into the plastic beta cell during or after radioactive counting. Using a small volume and low pressure range thermal conductivity sensor, the stable xenon behavior was measured before, during, and after radioactive counting. By monitoring the xenon concentration distribution as a function of time, xenon back diffusion to the sensor location as well as permeation into plastic beta-cells can be quantified. Accounting for these effects provides a more accurate estimate of the stable xenon concentration. Data from experimentation and modeling will be presented.

Log: 360. **A LEXICON FOR THE CONSISTENT DESCRIPTION OF SCANNING ELECTRON MICROSCOPY IMAGES FOR NUCLEAR FORENSICS.** Tamasi, A.L.(1,2); Eley, C.(3); Ross, A.R.(2); Ruggiero, C.E.(2); Porter, R.B.(2); Tandon, L.(2); Wagner, G.L.(2); Walensky, J.R.(1); Wilkerson, M.P.(2). (1) University of Missouri. (2) Los Alamos National Laboratory. (3) South Carolina State University.

Morphological analysis can provide a wealth of information about a sample, including details of potential process history, and as such is widely used in nuclear forensics in conjunction with trace chemistry, radiochemistry, and other methods of analysis. One of the most prominent methods for exploring morphology is Scanning Electron Microscopy (SEM), which can provide information about material type, surface texture, particle structure and size, and grain boundaries. However, there are no concrete numerical outputs from SEM, only an image, and it is difficult to objectively describe an image. Thus, image descriptors can vary widely from one researcher to the next, and it can be difficult to effectively search previously reported SEM data, and find images in the literature that are comparable to one's samples. Materials of interest can contain a wide variety of textures, shapes, and agglomerations, all of which could provide relevant information for nuclear forensic analyses. Bearing these issues in mind, we have designed a lexicon to consistently describe SEM data of nuclear forensics materials. We present the full lexicon along with representative drawings for each descriptor, and we detail step-by-step characterization of example materials using our lexicon. LA-UR-14-28792.

Log: 361. **PERFORMING FIELD CALIBRATIONS OF RADIOXENON DETECTORS.** Foxe, M.P.(1); Cameron, I.M.(1); Cooper, M.W.(1); Haas, D.A.(1); Hayes, J.C.(1); Hubbard, C.W.(1); Lidey, L.S.(1); McIntyre, J.I.(1); Mendez, J.M.(1); Prinke, A.M.(1); Riedmann, R.A.(1); Ripplinger, M.D.(1). Pacific Northwest National Laboratory.

Beta-Gamma coincidence radioxenon detectors must be calibrated for each of the four-radioxenon isotopes (Xe-135, Xe-133, Xe-133m, and Xe-131m). Without a proper calibration, there is potential for the misidentification of the amount of each isotope detected. It is important to accurately determine the amount of each radioxenon isotope, as the ratios can be used to distinguish between an anthropogenic source, and a nuclear explosion. We have developed a xenon calibration system (XECALS) that produces calibration spikes of known activity and pressure for field calibration of detectors. The specific activities of these calibration spikes are measured using a beta-gamma coincidence detector and a high purity germanium (HPGe) detector. We will present the results from the development and commissioning of XECALS, along with the future plans for a portable spike implementation system.

Log: 362. **DETECTOR EFFECTS ON THE MINIMUM DETECTABLE CONCENTRATION FOR RADIOXENON.** Foxe, M.P.(1); Cooper, M.W.(1); Day, A.R.(1); Haas, D.A.(1); Hayes, J.C.(1); McIntyre, J.I.(1); Prinke, A.M.(1). (1) Pacific Northwest National Laboratory.

Radioxenon detectors are used as part of the international monitoring system (IMS) for verification of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). We have studied the effects of cell design and performance on the minimum detectable concentration of Xe-135, Xe-133, Xe-133m, and Xe-131m. We have seen that the light collection of the beta cell varies with manufacturing process, impacting the MDC for each radioxenon isotope differently. Among identically constructed gas cells, configurations in passive shielding have been shown to cause changes in the MDC values. The effects of the gas purity from trace radon levels were investigated. In addition we performed a comprehensive study of the effect of gas collection time, sample count time, and xenon volumes for a variety of potential radioxenon detection systems. We will present the results of the studies and show the potential benefit of future systems.

Log: 363. **SIMULTANEOUS DETERMINATION OF MULTIPLE ACTINIDE ELEMENTS IN A VARIETY OF SOILS UTILIZING A STANDARDIZED SEQUENTIAL EXTRACTION PROTOCOL.** Faye, S.A. (1); Gallardo, A.M (1); Richards, J.M. (1); Sudowe, R. (1). (1) University of Nevada, Las Vegas.

To obtain a better understanding of the mobility of actinide element contaminants in the environment, the association of the contaminants with specific host phases of soils and sediments must be investigated. This investigation is most commonly conducted using sequential extraction techniques that employ the selective dissolution of soil phases with increasingly aggressive chemical treatment. Though several sequential extraction studies exist in literature, conclusions cannot be made by direct comparison of data due to the lack of uniformity in the protocols used. This work aims to continue investigations of the development of a standard sequential extraction protocol while also expanding capabilities to include the analysis of multiple actinide elements across a broad range of soil profiles. A five-step sequential extraction procedure was used to analyze extraction behaviors of <sup>232</sup>Th, <sup>238</sup>U, <sup>239,240</sup>Pu and <sup>241</sup>Am from four standard reference materials chosen to represent a broad range of geochemical soil profiles. Radioanalytical separations were performed using an extraction chromatography based separation procedure prior to CeF<sub>3</sub> microprecipitation sample mounting and counting by alpha spectroscopy. Stable element analysis of aluminum, iron, manganese, and strontium was performed by inductively coupled plasma – mass spectrometry to monitor phase selectivity and model potential fission product mobility. An investigation of complete dissolution techniques for a potential sixth fraction was conducted for the application to soils with high concentrations of insoluble metals. Results show variations in leaching behaviors potentially caused by geochemical host phase of the soil, chemical form of the extracted elements, and environmental weathering conditions.



Log: 364. **MULTI-ISOTOPE PROCESS (MIP) MONITOR DEPLOYMENT AT H-CANYON.** Meier, D.E.(1); Schwantes, J.M.(1); Casella, A.J.(1); Abrecht D.G.(1); Sexton, L.T.(2); Landsberger, S.(3); Coble, J.B.(4). (1) Pacific Northwest National Laboratory. (2) Savannah River National Laboratory. (3) University of Texas-Austin. (4) University of Tennessee-Knoxville.

Researchers at the Pacific Northwest National Laboratory in conjunction with Savannah River National Laboratory, University of Tennessee-Knoxville and the University of Texas-Austin are working to deploy the Multi-Isotope Process (MIP) Monitor at the H-Canyon Reprocessing Facility at the Savannah River Site. The MIP monitor is a radiation detection system coupled with multivariate analysis for monitoring spent nuclear fuel recycling facilities on-line, non-destructively, and in near-real-time. The goal of the deployment project is to gather and process the continual acquisition of the spectral data in the hot and warm sampling aisles at the H-Canyon reprocessing facility during actual reprocessing campaigns. The processed information could be used to improve safety, efficiency, material and process controls, and overall plant performance to aid both the regulator and the plant operator. This technology is intended to support the DOE Nuclear Energy's Fuel Cycle Research and Development (FCR&D) and Materials Protection, Accounting and Control Technology (MPACT) program. This talk will present an overview of the MIP Monitor deployment project, provide a status update, and show data from current deployment efforts.

Log: 365. **PLUTONIUM OXIDE PROCESSING PROJECT AT THE RADIOCHEMICAL PROCESSING LABORATORY.** Meier, D.E.(1); Tingey, J.M.(1); Lumetta, G.J.(1); Delegard, C.H.(1); Casella, A.J.(1); Rapko, B.M.(1); Edwards, M.K.(1); Orton, R.D.(1); Schwantes, J.M.(1); (1) Pacific Northwest National Laboratory.

Researchers at the Pacific Northwest National Laboratory (PNNL) in conjunction with other DOE national laboratories are developing a Laboratory Scale Plutonium Oxide processing capability for the purpose of improving our understanding of product material characteristics unique to specific processes. This capability at PNNL will be capable of producing 200 g of plutonium oxide quarterly under carefully controlled conditions. The effort will be conducted in the Radiochemical Processing Laboratory (RPL), which is a category-II non-reactor nuclear facility. The product material will be analyzed for unique physical and chemical characteristics. PNNL will also perform analytical characterization of the source material as well as material collected at various intermediate points for material control, accountability and characterization purposes. These product material "signatures" could then be used to support a variety of nuclear security efforts, including forensic analysis of interdicted plutonium. This project is supported by the National Technical Nuclear Forensics Center (NTNFC), a directorate within the Department of Homeland Security's Domestic Nuclear Detection Office. This talk will present an overview of the PuO<sub>2</sub> processing project and discuss radioanalytical, chemical and morphological capabilities at PNNL.

Log: 366. **PLATING OF IRIIDIUM FOR USE AS HIGH PURITY ELECTRODES IN THE ASSAY OF ULTRAPURE COPPER.** Mathews, Martin(1); LaFerriere, B. D.(1); Pederson, L. R.(1); Hoppe, E. W.\*(1). (1) Pacific Northwest National Laboratory (\*corresponding author).

The goal of this work was to fabricate high-purity electrodes via electrochemical deposition for use as anodes in the assay of ultrapure copper for uranium and thorium contamination primarily in support of the Majorana collaboration. High-purity iridium was deposited electrochemically on an iridium wire using a low current density under a constant voltage to achieve a thin film (about 1  $\mu\text{m}$ ) with a smooth surface. Current efficiency was as high as 23% using an electrolyte of 40 mM  $\text{IrCl}_3 \cdot 4\text{H}_2\text{O}$ , and sulfuric acid as the sole additive, direct current voltage without a reference electrode, and fine polished iridium wires as electrodes at an elevated temperature of 80 degrees C. Digital optical microscopy and scanning electron microscopy showed that a uniform and smooth surface was achieved. These pure iridium electrodeposited electrodes can be converted efficiently to iridium oxide with different oxidation states if desired by heating them in air for various periods. The resulting electrodeposited iridium electrodes are very promising for use in the analysis of high-purity copper in applications where radioactive contaminant species such as uranium and thorium must be kept to a minimum.

Log: 367. **STABILITY OF PLUTONIUM ASSOCIATED WITH CLAYS FROM HYDROTHERMALLY ALTERED NUCLEAR MELT GLASS.** Zavarin, M.; Begg, J.D.; Zhao, P.; Boggs, M.A.; Joseph, C.; Dai, Z.; Kersting, A.B. Glenn T. Seaborg Institute, Lawrence Livermore National Laboratory.

At the Nevada National Security Site (formerly Nevada Test Site), colloid-facilitated Pu transport has been identified as the dominant mechanism leading to downgradient migration of Pu. The Pu appears to be associated with clay and zeolite colloids. However, the nature of the association (e.g. surface adsorption, ion exchange, co-precipitation) has not been identified. We initially investigated Pu adsorption/desorption rates from montmorillonite colloids under controlled laboratory conditions. These data suggest that Pu desorption rates are slow enough that colloid-facilitated transport of adsorbed Pu may be possible at the field scale (km distances and decade timescales). However, an alternative scenario at the NNSS is Pu incorporation into secondary mineral colloids during nuclear melt glass alteration. This may lead to an irreversible association of Pu with colloids (e.g. co-precipitation). We investigated this mechanism through long-term hydrothermal alteration experiments. Nuclear melt glass was reacted for ~3 years at temperatures ranging from 25 to 200 degrees Celsius. Aqueous and colloidal Pu concentrations were monitored in the supernatant along with colloid concentrations/mineralogy and solid phase mineralogy. At the end of the 3-year experiment, Pu desorption was tested with batch extraction techniques. Preliminary data suggest that some Pu may partition into the alteration phases in such a way as to be inaccessible to desorption processes. This implies that colloid facilitated Pu transport may, in part, be controlled by the physical filtration processes that limit colloid transport rates rather than adsorption/desorption processes.

Log: 368. **ALPHA SPECTROSCOPY SUBSTRATES BASED ON ULTRATHIN POLYMER FILMS.** Mannion, J.(1); Husson, S.(1); Powell, B.(2); Locklair, D.(2). (1) Clemson University Department of Chemical and Biomolecular Engineering (2) Clemson University Department of Environmental Engineering and Earth Sciences. In this work, polymer thin film-coated glass slides were used as a novel substrate for the simultaneous concentration of actinides and sample preparation for alpha spectroscopy analysis. Detection of alpha emitting radioisotopes using alpha spectroscopy requires careful sample preparation to avoid self-absorption of the alpha particles, which can have deleterious effects on the resolution of the alpha spectrum. However, rapid methods, such as lanthanum fluoride microprecipitation, require hazardous chemicals (i.e. hydrofluoric acid). Thus, a new method was developed for the rapid, inexpensive, and efficient preparation of alpha spectroscopy samples that retains the high resolution of those prepared by electrodeposition or microprecipitation. Substrates were prepared by forming ultrathin films (<10 nm) of quaternary amine anion-exchange polymers on glass surfaces by dip-coating. Poly(vinylbenzyl chloride) films cross-linked with diazabicyclo[2.2.2]octane and activated through exposure to triethylamine were used for these studies; however, the general methodology developed can be extended to many other polymers bearing selective ligands. Synthesis of substrates is rapid and the methodology can be applied to virtually any substrate geometry and size. Samples were loaded by submerging the polymer-coated substrates into acidified solutions containing spiked <sup>238</sup>Pu or an acidified natural water with elevated uranium concentrations. The quality of the alpha spectra from these substrates indicates they will be a useful, inexpensive, and potentially field deployable platform serving national security and environmental sampling applications.

Log: 369. **A New Instrument for Cold Neutron Prompt Gamma-ray Activation Analysis at NIST.** Rick L. Paul (1), Dagistan Sahin (1), Christoph Brocker (2), Jeremy C. Cook (2), Richard M. Lindstrom (1), Donna J. O'Kelly (1). (1) National Institute of Standards and Technology, Chemical Sciences Division. (2) National Institute of Standards and Technology, Center for Neutron Research.

A new spectrometer for cold neutron activation analysis is now operational at the NIST Center for Neutron Research (NCNR). The new instrument on NGD guide has higher neutron flux, lower background, and greater applicability than a previous instrument at NG7. A beam of cold neutrons flows through a lithoflex-lined beam tube into a  $^6\text{Li}$  glass-lined sample chamber. The chamber and beam tube are evacuated, reducing background from air scattering and from hydrogen and nitrogen in air. Fast neutrons, produced by the secondary reactions of beam neutrons with  $^6\text{Li}$ , are ameliorated with borated polymer shielding surrounding the instrument, reducing risk of damage to the detector. Prompt gamma rays are measured by a lead-shielded germanium detector with a bismuth germanate Compton shield. Signals are processed using a pair of digital signal analyzers. A thermal equivalent neutron flux of  $6.8\text{E}9\text{ cm}^{-2}\text{s}^{-1}$  is measured at the sample position, a factor of 10 higher than at NG7. Hydrogen background is a factor of  $\approx 5$  lower than at NG7, with background from other elements similarly reduced. The limit of detection for hydrogen is on the order of  $< 100\text{ ng}$ . A low background two detector coincidence/add-back counting system for determination of decay gamma rays, located a few feet from the instrument, further enhances the applicability of the facility. Future improvements will include the addition of a scanning stage, sample changer, sample chambers for studying in situ reactions, and additional detectors for coincidence prompt gamma measurements.

Log: 370. **DEVELOPING RADIUM-226 AND ACTINIUM-227 AGE-DATING TECHNIQUES FOR NUCLEAR FORENSICS TO GAIN INSIGHT FROM CONCORDANT AND NON-CONCORDANT RADIOCHRONOMETERS.** Kayzar, T.M. (1); Williams, R.W. (1). (1) Lawrence Livermore National Laboratory.

The model age or 'date of purification' of a nuclear material can be an important nuclear forensic signature. Age determination, however, relies on fundamental assumptions that (1) at time  $t = 0$ , the parent nuclide was completely purified, and (2) the material remained a closed system following purification. These assumptions may not be valid for all nuclear material. In this study, we developed separation and measurement techniques for radium-226 and actinium-227: grand-daughter nuclides in the decay chains of uranium-238 and uranium-235 respectively. The behavior of grand-daughter nuclides in nuclear material can provide additional information in forensic cases where pairs of parent-daughter model ages are discordant or inconsistent with the known history of the material. Enriched  $^{228}\text{Ra}$  and  $^{229}\text{Th}$  ( $^{225}\text{Ac}$ ) tracers were purified and calibrated using the NIST4967A  $^{226}\text{Ra}$  standard and Ames  $^{232}\text{Th}$  metal. Isotope dilution  $^{226}\text{Ra}$  concentrations were measured using the  $^{228}\text{Ra}$  tracer for: CRM U100 – purified in January 1959; Table Mountain Latite - a geologic matrix with U-series decay ( $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ - $^{226}\text{Ra}$ ) in secular equilibrium; ores certified for Ra activity by CANMET; and two HEU metal logs used for the International Technical Working Group Round Robin 3 Exercise. Multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS, Nu Plasma HR) measurement techniques were developed to measure isotope ratios  $^{226}\text{Ra}/^{228}\text{Ra}$  and  $^{227}\text{Ac}/^{225}\text{Ac}$ . Accurate Ra concentration measurements are reported for samples with only hundreds of femtograms of  $^{226}\text{Ra}$ . Isotope dilution  $^{227}\text{Ac}$  concentrations of  $^{235}\text{U}$ -enriched nuclear materials were also measured. Model ages calculated from grand-daughter chronometers will be discussed in relation to parent-daughter model ages. LLNL-ABS-665333.

Log: 371. **NEW REFERENCE MATERIALS FOR TRACE-LEVEL ACTINIDE ELEMENTS IN URANIUM OXIDE.** Essex, R.M. (1), Williams, R.W. (2), Gaffney, A.M. (2), Hexel, C.R. (3), Bostick, D. (3), Morales, M. (4), Croatto, P. (1), Mathew, K.J. (1), Morrison, J.J. (5). (1) New Brunswick Laboratory. (2) Lawrence Livermore National Laboratory. (3) Oak Ridge National Laboratory. (4) Savannah River National Laboratory. (5) Department of Homeland Security.

NFRM U-1 and NFRM U-2 are new analytical reference materials for characterization of trace-level actinide elements in a uranium oxide powder. This project was sponsored by the Department of Homeland Security (DHS) and was a cooperative effort between New Brunswick Laboratory, Lawrence Livermore National Laboratory (LLNL), and Oak Ridge National Laboratory (ORNL). Each unit of NFRM U-1 and NFRM U-2 is comprised of 1.5 grams of U3O8 powder in a 20-mL glass scintillation vial. The oxides are highly enriched in U-235 (93% for U-1 and 52% for U-2) and were previously certified for U isotope amount ratios and U amount content as the base materials for a  $\gamma$ -spectrometry reference material, CRM 146. For this project, multiple subsamples of oxide were dissolved and processed to determine the trace concentrations of Th, Np, Pu, and Am. Isotope amount ratios were also determined for the Th and Pu trace constituents. Analyses of the Np and Am in the oxides indicated that these elements are mono-isotopic. The characterization data from LLNL were used to determine attribute values for the reference materials. Verification results from ORNL and additional analytical results from labs participating in a DHS-sponsored methodology benchmarking study were used to assess accuracy, reproducibility, and measurement uncertainties. Expanded uncertainties for the attribute values are compliant with international standards and characterization measurements are traceable to the U.S. national reference base. The attributes in the oxides are sufficiently homogeneous and reproducible to serve as certified reference materials for trace actinide element content in a U oxide.

Log: 372. **IMPROVEMENTS IN TELLURIUM ISOTOPE MEASUREMENTS: IMPLICATIONS FOR NUCLEAR FORENSICS.** Shollenberger, Q.R.(1); Brennecka, G.A.(1,2); Ramon, E.C.(1); Borg, L.E.(1); Hutcheon, I.D.(1). (1) Lawrence Livermore National Laboratory. (2) University of Münster.

The ability to recognize identifiable characteristics – such as isotopic signatures – in nuclear material is imperative to the nuclear forensics community. Certain isotopic systems, such as the fission product tellurium (Te), have largely been ignored in part due to difficulties associated with precise isotopic measurements. Previous studies have measured Te isotopes using traditional MC-ICPMS and N-TIMS [1, 2] with insufficient precision to answer many interesting questions. The common challenges hindering high-precision measurement of this isotopic system include: a high first ionization potential to forming positive Te ions, numerous isobaric interferences, and the low natural abundance of Te in the samples of interest. However, recent work has shown that the instrumental sensitivity for elements, that readily form a hydride, like Te, can be significantly increased by introducing the sample as a hydride gas to the ICPMS [3]. Using hydride generation, we report an increase in sensitivity of greater than 50× with overall precision improved by more than 30× when compared with traditional MC-ICPMS. Additionally, hydride generation decreases the potential for matrix induced interferences by preventing non-hydride forming elements from entering the plasma. These advances represent a significant improvement to current measurement capability and open Te isotopes as a promising new signature in nuclear material. References: [1]Fehr et al. (2009) MAPS, 44, 971. [2]Fukami et al. (2014) JAAS, 29, 520. [3]Forrest et al. (2009) Geostandards and Geoanalytical Research, 33, 261. Prepared by LLNL under contract AC52-07NA27344. Document LLNL-ABS-664761.

Log: 373. **CAPTURING 3D MORPHOLOGY OF NUCLEAR FORENSIC SPECIMENS IN SUB-MILLIMETER RESOLUTION USING STEREOPHOTOGRAMMETRY SOFTWARE AND A DIGITAL CAMERA.** Robel, M. (1), Knight, K. B. (1), Kristo, M. J. (1), Tryshyn, V. (2) and Malyuk, I. (2). (1) Lawrence Livermore National Laboratory, Livermore, USA. (2) Kyiv Institute for Nuclear Research, Kyiv, Ukraine.

Stereophotogrammetry methods are used in diverse applications from crime scene reconstruction to paleontology to generate 3D models from an array of photographs. In this study, we explore the application of these methods to nuclear forensic analysis of interdicted uranium oxide fuel pellet fragments for both qualitative and quantitative three-dimensional specimen characterization. Comparative nuclear forensic analysis of unknown specimens seeks to rule in and rule out possible sources through comparison to known samples. When possible, prior to destructive analysis, nuclear forensic specimens are characterized by non-destructive means for physical parameters including mass, geometry, volume, and density. Stereophotogrammetry offers a method to capture additional information for physical sample characterization in the form of high-precision, 3-D digitized models of specimens. These 3-D models can be used quantitatively to determine volume (for density determination), as well as a variety of sample dimensions that may be difficult to measure directly or through optical images, alone. Such models can also be used to generate a virtual reconstruction of the possible original object, when only fragments of a sample are available for analysis. We will illustrate these capabilities using fragments of interdicted fuel pellets. Stereophotogrammetry tools are relatively inexpensive and require minimal operator training, making these methods accessible for any level of nuclear forensics capability. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory (under contract DE-AC52-07NA27344). LLNL-ABS-665328



Log: 374. **APPLICATION OF NEUTRON ACTIVATION ANALYSIS TO MICRO GRAM SCALE OF SOLID SAMPLES.** Sekimoto, S.(1); Okazaki, R.(2); Itosu, S.(1); Hirayama, Y.(2); Tobimatsu, Y.(2); Ohtsuki, T.(1). (1) Kyoto University Research Reactor Institute. (2) Kyushu University.

Neutron activation analysis (NAA) is non-destructive and has frequently been used for bulk elemental analysis of unique samples, such as special phases collected from meteorites or extraterrestrial dust particles. Chondritic and iron meteorites contain relatively high contents of Co and Ir compared with those in the earth crust and differentiated stone meteorites. As Co and Ir have high sensitivities in NAA, they can be good markers for the identification of such extraterrestrial materials. In NAA of chondrites, a few tens mg of specimen is commonly used. For such a case, a few ng of Ir and a few micro-g of Co can be reliably determined. When an extremely small-sized (i.e., less than micro gram) samples, such as micrometeorites, IDPs (Interplanetary Dust Particles), and dust particles returned from asteroids or comets, are to be analyzed by NAA, the conventional NAA procedure used for a few tens mg is not suitable. For such tiny samples, neutron irradiation with high neutron flux and long irradiation time is required. For the irradiation with high neutron dose, polyethylene bags for holding samples are not usable because they are prone to radiation damage. Polyethylene bags are also not suitable for holding tiny grain samples. It is, therefore, very important to design and improve an appropriate sample holder for irradiating small grain samples. In this study, we present the NAA procedure for analyzing a single grain of down to micro and sub-micro grams in mass. As we use a relative method for quantification, the preparation of reference samples and the evaluation for their suitability are highly concerned and, therefore, discussed in detail.

Log: 375. **DIFFUSIVE MASS TRANSPORT OF FE, CA, MG, AND U, IN AERODYNAMIC FALLOUT GLASS FROM A NEAR-SURFACE NUCLEAR EXPLOSION.** Weisz, D.G.(1); Prussin, S.G.(1); Knight, K.B.(2); Marks, N.E.(2); Jacobsen, B.(2); Hutcheon, I.D.(2). (1) University of California, Berkeley. (2) Lawrence Livermore National Laboratory.

In the fireball of a near-surface nuclear explosion, surrounding soil and device material mix together. During cooling of the fireball, the melted soil quenches, often forming symmetric glassy objects known as aerodynamic fallout glass. The conditions and chemical processes through which device material and soil interact to form these objects are not well understood. However, these details are critical to fallout formation modeling, which is intended to improve sample interpretation for post-detonation nuclear forensic analysis. The goal of this study is to constrain the time scales and temperature ranges that control mass transport in aerodynamic fallout glasses by studying compositional variation in these objects. The fallout glasses in our study were obtained from a historical US test and consist of millimeter-scale objects with spheroidal, sub-millimeter sized objects attached to their surfaces. The compositional variations of major and minor elements were measured using energy and wavelength dispersive x-ray spectroscopy. Uranium concentration and isotopic variation were measured using nano-secondary ion mass spectrometry (NanoSIMS). The interfaces between the small objects and the larger host objects are consistently enriched in iron, calcium, magnesium, and uranium. The concentration profiles of these elements suggest chemical diffusion. While the effect of composition and temperature variation across the interfaces need to be better understood, and improved data on diffusivities in rhyolitic glasses are needed, our results provide compelling evidence that the time and temperature of mass transport can be constrained by studying chemical and self-diffusion profiles in these materials. Prepared by LLNL under Contract DE-AC52-07NA27344. LLNL-ABS-664935.

Log: 376. **DEVELOPMENTS IN ISO STANDARDS FOR RADIONUCLIDE METROLOGY.** Judge, S.M.(1); Jerome, S.M.(1). National Physical Laboratory, UK.

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies. Measurement laboratories refer to ISO standards for instructions and guidance on determining the radioactivity content of samples (use of these standards is compulsory for laboratories seeking accreditation to ISO17025); as such, ISO standards are an important mechanism to promulgate best practice in radionuclide metrology. The work of preparing such international standards is normally carried out through ISO technical committees. This presentation will give an overview of recent developments in ISO standards for topics such as gamma spectrometry and the measurement of surface contamination. The new approach to harmonising the standards will be discussed and the contribution made by National Measurement Institutes through the technical committees will be highlighted.

**Log: 377. DIRECT ELECTRON IRRADIATION OF THE URANYL SULFATE SOLUTION: BUBBLE FORMATION AND THERMAL HYDRAULICS STUDIES.**

Sergey Chemerisov, Roman Gromov, Vakho Makarashvili, Thad Heltemes, Zaijing Sun, Kent Wardle, James Bailey, Dominique Stepinski and George Vandegrift Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439.

Argonne is assisting SHINE Medical Technologies in developing SHINE, a system for producing Mo-99 with accelerator-driven subcritical target solution of uranyl sulfate. In order to simulate conditions expected in SHINE, an experimental setup based on LINAC accelerator was built at Argonne to study radiolysis, thermal-hydraulics, and bubble formation in the uranyl sulfate solution. The target bubble chamber is cooled by the front and back surfaces and by a central tube to mimic the geometry of the proposed SHINE solution vessel. Multiple thermocouples are inserted into solution to map its thermal profiles. An electron beam from the LINAC accelerator were rastered to irradiate 20L uranyl sulfate solution homogenously. The thermal-hydraulic model based on OpenFoam and radiation model based on MCNPX are tested and verified by the experiments.

**Log: 378. URANIUM ISOTOPE CONSTRAINTS ON CONTAMINANT SOURCES IN TREEBARK.** Conte, E.R.; Widom, E. ; Kuentz, D. Miami University, Oxford OH.

Environmental materials (i.e. tree bark, lichen) can record local atmospheric pollution. Uranium (U) isotopic analysis of these materials can provide a means to distinguish between the presence of natural and non-natural sources of U. The former Fernald Feed Materials Production Center (FFMPC; Ross, Ohio) produced U metal from natural U ores and recycled nuclear materials from 1951-1989, and it is estimated that the FFMPC accidentally released ~90-450 Mg of U into the environment [1]. Alba Craft Laboratory (Oxford, Ohio) machined several hundred tons of nominally natural U metal from FFMPC between 1952-1957, resulting in significant environmental U contamination. We have measured U concentrations and isotopic ratios in tree bark samples from a NNW-SSE transect between the Alba Craft Laboratory site and the FFMPC to investigate the potential of tree bark as an effective biomonitor. Our results demonstrate the presence of anthropogenic U contamination in tree bark from the entire study area, with U concentrations at Fernald up to ~575X background levels. Samples exhibit  $^{235}\text{U}/^{238}\text{U}$  values from 0.00461 to 0.00725, becoming progressively more depleted towards the FFMPC [this study & 2,3]. Additionally,  $^{236}\text{U}/^{238}\text{U}$  increases progressively towards the FFMPC, with values as high as  $2.00\text{E-}4$ . 1. Makhijani, A., 2000. SDA. 5. 2. Widom, E. and Kuentz D., 2010. GSA abstract, Denver. 3. Conte and Widom. 2013. GSA abstract, Denver.

Log: 379. **THE ROLE OF ORGANIC SOLVENT RADICAL CATIONS IN SEPARATIONS LIGAND DEGRADATION.** Mezyk, S.P. (1); Mincher, B.J. (2); Cook, A.R. (3); Wishart, J.F. (3) (1) California State University at Long Beach (2) Idaho National Laboratory (3) Brookhaven National Laboratory.

Solvent based extractions using designer organic complexing agents have been the most commonly used process-scale separations technique for nuclear applications over the past 60 years. However, these extraction systems will be deployed in highly radioactive environments, and the radiation chemistry of these ligands and their diluents will play a major role in determining extraction efficiency, separation factors, and solvent-recycle longevity. Considerable work has been reported on radiation-induced ligand damage for acidic water radiolysis species. However, there are far less fundamental kinetic and mechanistic data available for the degradation of extraction ligands in the organic phase. The organic phase typically uses n-alkanes such as dodecane, TPH, and kerosene as diluents, and their radiolysis produces a mixture of radical cations ( $R^{\bullet+}$ ), carbon-centered radicals ( $R^{\bullet}$ ), solvated electrons and molecular products including hydrogen. Typically, radical species will preferentially react with dissolved oxygen, producing relatively inert peroxy radicals. This isolates the alkane radical cation species,  $R^{\bullet+}$ , as the major radiolytically-induced organic species that can react with, and degrade, extraction agents in this phase. Here we report on recent investigations of dodecane radical cation reactions with nitrogen-containing ligands. Fast kinetics were observed, stressing the importance of this mechanistic pathway. Elucidating these parameters, and combining them with known acidic aqueous phase chemistry, will enable a full, fundamental, understanding of the impact of radiation on solvent extraction-based separation processes. Work at the BNL LEAF Facility was supported by the U.S. DOE Office of Science, Division of Chemical Sciences, Geosciences, and Biosciences under contract DE-AC02-98CH10886.

Log: 380. **ACCELERATOR MASS SPECTROMETRY AT PRIME LAB: NEW TECHNIQUES FOR ULTRA-SENSITIVE MEASUREMENT OF RADIONUCLIDES.** Caffee, M.W. (1); Woodruff, T.(1). (1) Purdue University.

Accelerator Mass Spectrometry (AMS) enables the measurement of long-lived radionuclides such as  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ , and  $^{129}\text{I}$ . These radionuclides are produced by cosmic ray interactions with terrestrial and extra-terrestrial materials; some of these are also produced by nucleogenic processes. The measurement of these radionuclides in some instances requires the separation of the radionuclide of interest from isobaric interferences. This is typically accomplished with a  $dE/dx$  detector, but in many instances the radionuclide of interest cannot be resolved from the more abundant isobar. At PRIME Lab we are now routinely using a gas-filled-magnet (GFM) for the measurement of  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ , and  $^{36}\text{Cl}$ . In the near future we anticipate measurements of  $^{41}\text{Ca}$  and  $^{53}\text{Mn}$ . For all radionuclides measured to date the GFM does suppress isobaric interferences, allowing low backgrounds for samples with relatively high isobar concentrations. For  $^{26}\text{Al}$  we can now inject  $\text{AlO}$ , which produces a factor of  $\sim 10$  more current than  $\text{Al}^-$ . The higher beam currents directly translate into more counts at the detector. We can now measure  $^{26}\text{Al}/^{27}\text{Al}$  ratios with a precision comparable to  $^{10}\text{Be}/^9\text{Be}$  ratios.

Log: 381. **STUDIES OF EXTRACTABLE ORGANOHALOGENS IN FARMED ATLANTIC SALMON TISSUES BY NEUTRON ACTIVATION, VARIOUS CHROMATOGRAPHIC, NUCLEAR MAGNETIC RESONANCE AND MASS SPECTROMETRIC TECHNIQUES.** Bahroun N.H.O.; Chatt, A. Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, 6274 Coburg Road, Room 212, P.O. Box 15000, Halifax, NS, B3H 4R2, Canada.

Human exposure to persistent lipophilic halogenated organic compounds such as dioxins, PCBs, PBBs, etc. occurs mainly via food, especially fish. Salmon is considered to be a healthy food due to its high omega-3 fatty acid content. However, organohalogen contaminants are of major concern in this type of fatty fish. A detailed study of extractable organohalogen (EOX) in farmed Atlantic salmon tissue and skin samples was carried out in this work. An acetone:hexane (1:1) mixture was used for the extraction of halogens. An instrumental neutron activation analysis method in conjunction with anticoincidence (INAA-AC) gamma-ray spectrometry was used for the determination of halogens using the Dalhousie University SLOWPOKE-2 reactor (DUSR) facility. The detection limits of 4.4, 0.26, and 0.16 ng/g were obtained in tissues for chlorine, bromine and iodine, respectively. Size exclusion, solid-phase extraction and thin layer chromatographic analyses showed that both tissue and skin fractions had triacylglycerols. Then <sup>31</sup>P-, <sup>1</sup>H- and <sup>13</sup>C-NMR, positive-ion electrospray ionization and tandem MS/MS, and INAA-AC proved that most of the EOX was present in these triglyceride fractions. Attempts were then made by to deduce the composition of the individual triacylglycerides and investigate the presence of halogenated fatty acids in them.

Log: 382. **ENVIRONMENTAL CONTAMINATIONS WITH RADIOCESIUM IN LAKE ONUMA ON MT. AKAGI IN GUNMA PREFECTURE EMITTED BY THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT ACCIDENT.** Tsunoda, K.(1); Aizawa, S.(1); Mori, M.(1); Saito, Y.(1); Kozaki, D.(1); Koike, Y.(1); Abe, S.(1); Suzuki, K. (2); Kuge, T. (2); Izumi, S. (2); Tanaka, H. (2); Onozeki, Y. (2); Nohara, S (3); Minai, S (3); Okada, Y. (5); Nagao, S. (6). (1) Gunma University. (2) Gunma Prefectural Fisheries Experiment Station. (3) National Institute for Environmental Studies. (4) Musashi University. (5) Tokyo City University. (6) Kanazawa University.

FDNPP accident has resulted in contamination of the environment in Gunma Prefecture with radioCs (Cs-134 and Cs-137). In particular, concentrations of radioCs greater than 500 Bq/kg were found in wakasagi (*hypomesus nipponensis*) in Lake Onuma at the top of Mt. Akagi in August 2011. The contamination level remains high with radioCs concentrations greater than 90 Bq/kg being observed in November 2014. To elucidate the mechanism of this contamination, monitoring studies on Lake Onuma and other lakes and rivers in the prefecture were performed including analyses of fish, aquatic plants, plankton, lake and river water samples, lake sediments and soil samples together with hydrological investigations. The total concentrations for radioCs in Lake Onuma decreased gradually with time, but values were still more than 10 times higher than those for other lakes and rivers in the prefecture. Moreover, a high correlation was found between total concentrations of radioCs in lake waters and those in wakasagi. Considering the results for isotopic analyses of nitrogen and carbon in the food chain, the order for the food chain with respect to wakasagi in Lake Onuma is the order of lake water, phytoplankton, zooplankton, and wakasagi. The bioaccumulation factor for radioCs in wakasagi was about 1400. The mass balance and residence time for radioCs in Lake Onuma were also investigated. The outflow for radioCs from the lake was almost the same as the decrease of radioCs in the lake. Moreover, the radioisotope was estimated to circulate about 14 times per year in the lake.

Log: 383. **COOPERATION IN EDUCATION AND TRAINING IN NUCLEAR- AND RADIOCHEMISTRY IN EUROPE -A CONTINUOUS PROCESS.** T. Retegan(1),C. Ekberg(1), J. John(2), V. Cuba(2), M. Nemec (2), J. Lehto(3), T. Koivula (3), P.J. Scully (4), C. Whalters (5), N. Evans (6), L. Skipperud (7), B. Salbu (7), E. Ansoborlo (8), J.P. Omtvedt (9). (1) Chalmers, Sweden. (2)CTU, Czech Republic. (3)UH, Finland. (4)NNL,UK. (5)IRS, Germany. (6)LU, UK. (7)UMB, Norway. (8)CEA, France. (9) UiO, Norway.

The skills in nuclear chemistry are of strategic, as well as immediate, importance for the maintenance of European nuclear operations. The demand for these skills would not decrease even if Europe decides to phase out its nuclear energy because they are even more indispensable for decommissioning the nuclear installations than for their operation, and a substantial demand for these skills exists in non-energy sectors. A sustained effort has been put on identifying both the existing training and education capabilities in Europe (not only EU), including Russia as well as the actual demand on education in Nuclear and Radiochemistry from the End Users. The goal is to fill in the gaps and contribute actively to the existing efforts, according to the IAEA Systematic Approach for Training (SAT), where the implementation have been conducted in two successive projects: Analysis, Design and Development (CINCH-I) and Implementation and Evaluation (CINCH-II). The outcomes as well as the continuous effort carried-out on the direction of fulfilling the coherence of education and training on Nuclear and Radiochemistry in Europe will make the subject of this work.

Log: 384. **EFFECT OF DESORPTION AND REDOX KINETICS ON RADIONUCLIDE TRANSPORT IN CRYSTALLINE FORMATIONS.** Schäfer, T. (1); Huber, F.M. (1); Lagos, M.(1); Quinto, F.(1); Heck, S. (1); Martin, A. (2); Blechschmidt, I.(2). (1) Institute for Nuclear Waste Disposal (INE), KIT. (2) NAGRA, Wettingen, Switzerland. Transport of radionuclides in crystalline environments might occur through dissolved species or attached to colloidal or nanoparticulate phases being mobile in the water conducting features of the host rock. In this presentation we discuss observed Tc-99, U-233, Np-237, Pu-242 and Am-243 sorption/desorption kinetics with and without natural or synthetic clay minerals (smectites) under conditions expected in the evolution of crystalline groundwater systems. The desorption or redox kinetics were monitored over a duration of up to 10,000h using natural fracture filling material as a concurrence ligand and monitoring the colloid attachment via detection of Al, Si, Ni and Zn as smectite structural elements. For trivalent actinides smectite desorption rates in the range of  $1.2\text{--}3.7\text{E-}3$  per hour could be determined and significantly lower desorption rates for tetravalent actinides were found. This results will be compared with laboratory core migration studies and field data on migration experiments performed at the Grimsel Test Site (GTS, Switzerland) using the same radionuclides and colloidal phases varying the fracture residence time by flow rate adjustment. Furthermore, the long-term actinide mobility will be addressed by presenting AMS/RIMS measurements of (a) samples collected several months into the tailing of the breakthrough curves not any longer detectable by HR-ICP-MS and (b) background samples of different GTS ground waters showing fallout U-236, whereas fallout Pu could not be detected indicating a much lower mobility under the given conditions.

Log: 385. **ASSESSING ENVIRONMENTAL ARCHIVES OF THE SOURCE AND DISTRIBUTION OF URANIUM CONTAMINATION.** Widom, E.(1); Kuentz, D.C.(1). (1) Miami University, Oxford OH. Our research group has been investigating uranium concentrations and isotope ratios in a variety of biomonitors (tree bark, lichen and tree rings) and pond sediment to establish their utility as archives of past uranium releases to the environment for purposes of environmental remediation and nuclear forensics. Our studies have focused on the area of the former Fernald Feed Materials Production Center, for which there are historical records estimating releases to the environment of 90-450 Mg of uranium during its years of operation (1951-1989). Our results demonstrate that tree bark and lichen retain signatures of past atmospheric dispersal of uranium including DU and EU, and that they are ideal archives for mapping the aerial extent and locating the source of uranium contamination. Our studies have further demonstrated that pond sediment can retain a temporal record of uranium contamination, and that variations in isotopic ratios ( $^{235}\text{U}/^{238}\text{U}$ ,  $^{236}\text{U}/^{238}\text{U}$  and  $^{234}\text{U}/^{238}\text{U}$ ) record changes through time from emission of DU to EU. The presence of measureable  $^{236}\text{U}$  in the biomonitors and sediment serves as a particularly sensitive tracer of non-natural uranium even when  $^{235}\text{U}/^{238}\text{U}$  ratios are within measurement error of natural values, thus  $^{236}\text{U}/^{238}\text{U}$  measurements are key for forensic applications.



Log: 386. **EFFECT OF FERROUS IRON POOL AND MAGNETITE STOICHIOMETRY ON Tc(VII) AND U(VI) RETENTION.** Totskiy, Y. (1); Huber, F.M. (1); Schild, D. (1); Heck, S. (1); Geckeis, H. (1); Kalmykov, S. (2); Schäfer, T.(1). (1) Institute for Nuclear Waste Disposal (INE), KIT. (2) Lomonosov Moscow State University, Moscow, Russia.

Information on Tc mobility under conditions similar to the host rocks for deep geological nuclear waste repositories is necessary for safety assessment during the repository development. This work deals with the interaction of Tc(VII) with crystalline rock materials from the generic underground research laboratory in Sweden (Äspö Hard Rock Laboratory) and from a prospective site of nuclear waste and SNF disposal in Russia (Nizhnekansky massif). Drilling of the Äspö cores was performed under anoxic conditions to preserve in-situ conditions. It was found, that Tc(VII) immobilization on the crystalline rocks is strongly dependent on sample preservation conditions. This results can be explained by Tc(VII) reduction to the insoluble Tc(IV) oxide by the ferrous iron pool available in the granite. Tc(VII) reduction was proved by XPS and XANES analyses of granite surface after sorption. Additional studies on synthetic magnetite revealed a strong dependency of Tc(VII) and U(VI) reduction on the magnetite stoichiometry with reduction reactions diminishing at a Fe(II)/Fe(tot) ratio of approx. 0.2 determined by XPS. Rs values for the both oxidized ÄD and NK materials are almost similar. Desorption of radionuclide is insignificant under natural conditions, but after artificial oxidation of samples technetium mobility is significantly increased. To investigate the Tc mobility under near-natural conditions an un-oxidized ÄD core with a natural fracture was used for column migration studies. Tc migration is investigated using Tc-95m isotope with concentration of ~10-11 M. Tc recovery is inversely dependent on residence time showing significantly increased reduction kinetics compared batch sorption studies.

Log: 387. **INFLUENCE OF POREWATER INJECTION ON ESTUARINE MIXING OF SOLUTES IN THE JIULONG RIVER ESTUARY, SOUTHEAST CHINA: A 224RA/228TH DISEQUILIBRIUM METHOD.** Hong, Q.Q.(1,2); Cai, P.H.(1,2); Shi, X.M.(1,2); Li, Q.(1,2); Liu, L.F.(1,2). (1) State Key Laboratory of Marine Environmental Science, Xiamen University. (2) College of Ocean and Earth Sciences, Xiamen University.

Here, we utilize the newly developed 224Ra/228Th disequilibrium approach to determine the benthic fluxes of solutes (DIC, NH<sub>3</sub>-N, NO<sub>x</sub>-N, PO<sub>4</sub><sup>3-</sup> and Mn) in the Jiulongjiang estuary, and to assess the influence of sedimentary input on the mixing behavior of these species. Benthic fluxes of 224Ra were calculated with a 1 D mass balance model. We demonstrated that irrigation dominates the benthic transfer of solutes. Based on the mass balance of water column 224Ra, site-specific residence times were estimated to be -44.26 to 2.75 d. The negative values were derived in the mid-estuary region, presumably resulting from our assumption of a negligible submarine groundwater discharge. By reconciling the water residence times, we were able to estimate the contribution of SGD to the area. We estimated benthic fluxes of DIC, NH<sub>3</sub>-N, PO<sub>4</sub><sup>3-</sup> and Mn to be -117.5–6236 mmol/m<sup>2</sup> d, 74.0–510 mmol/m<sup>2</sup> d, 0.94–27.0 mmol/m<sup>2</sup> d, and -22.1–137 mmol/m<sup>2</sup> d, respectively. On the contrary, bottom sediments removed NO<sub>x</sub>-N at a rate of -2195–56.6 mmol/m<sup>2</sup> d. The average benthic fluxes of DIC and PO<sub>4</sub><sup>3-</sup> over the whole estuary are approximately 160 mmol/m<sup>2</sup> d and 0.61 mmol/m<sup>2</sup> d. The DIC flux is comparable to the net export by SGD, whereas the PO<sub>4</sub><sup>3-</sup> flux is about two orders higher than the SGD-derived flux. In addition, they are equivalent to ~43% and ~105% of the riverine input. Our results highlight the important role of porewater injection in the water column budget of solutes in an estuary.

Log: 388. **USING  $^{224}\text{Ra}/^{228}\text{Th}$  DISEQUILIBRIUM TO QUANTIFY BENTHIC FLUXES OF DISSOLVED INORGANIC CARBON AND NUTRIENTS INTO THE PEARL RIVER ESTUARY.** Cai, P.H.(1,2); Shi, X.M.(1,2); Moore, W.S.(3); Hong, Q.Q.(1,2); Li, Q.(1,2); Guo, X.H.(1,2); Dai, M.H.(1,2). (1) State Key Laboratory of Marine Environmental Science, Xiamen University. (2) College of Ocean and Earth Sciences, Xiamen University. (3) Department of Earth and Ocean Sciences, University of South Carolina.

In this study, we utilize a newly developed approach – the  $^{224}\text{Ra}/^{228}\text{Th}$  disequilibrium approach, to quantify the benthic fluxes of dissolved inorganic carbon (DIC) and nutrients in a major estuary in China – the Pearl River Estuary (PRE). We demonstrate that irrigation was the predominant process that controls solute transfer across the sediment-water interface, whereas molecular diffusion and sediment mixing together accounted for <5% of the solute flux from bottom sediments. Based on the mass balance of water column  $^{224}\text{Ra}$ , an independent approach was proposed to estimate site-specific residence times of water mass. The results show that water residence times ranged from  $0.7 \pm 0.1$  to  $4.9 \pm 1.1$  d in the PRE. We show that sediment interstitial waters released approximately  $91.7 \pm 13.8 \times 10^9$  mol of DIC and  $\sim 37.1 \pm 5.6 \times 10^9$  mol of  $\text{NH}_4^+$  into the PRE in the dry season. On the contrary, bottom sediments removed  $\sim 15.7 \pm 1.3 \times 10^9$  mol of  $\text{NO}_2^- + \text{NO}_3^-$  from the overlying water column in this season. Overall, our results unravel porewater injection as an important process that controls the concentration levels of water column DIC and nutrients in estuaries. This study also highlights the  $^{224}\text{Ra}/^{228}\text{Th}$  disequilibrium approach as a very reliable tool for quantifying the rate of solute transfer across the sediment-water interface.

Log: 389. **Impact of atmosphere on the transport of Ruthenium in the primary circuit of nuclear power plant.** Kajan, I. (1); Kärkelä T.(2); Tapper, U.(2); Gouëlle, M.(2); Ramebäck, H.(1); Auvinen, A.(2) (1) Chalmers University of Technology. (2) VTT Technical Research Centre of Finland.

Ruthenium is a semi-volatile element existing as a fission product in nuclear reactor fuel that can be released in case of a severe nuclear accident. This release is promoted by air ingress, high humidity, high temperature and oxidative conditions in the reactor containment when the consistency of primary circuit is lost. In this work, the impact of gaseous atmosphere composition on the transport of ruthenium through a model primary circuit was examined. The first experiments were conducted in an air atmosphere. In the following experiments, additional silver nanoparticles were used to simulate aerosols transporting in the primary circuit. Furthermore, the impact of  $\text{NO}_2$  gas, as a product of air radiolysis, was also examined. The amount of ruthenium transported both as gas ( $\text{RuO}_4$ ) and aerosol was quantified. Chemical composition of ruthenium species was evaluated by XPS and Raman spectroscopy techniques. The transport of gaseous ruthenium through the model primary circuit increased significantly, when  $\text{NO}_2$  gas was mixed with the airflow. Addition of both silver aerosol and  $\text{NO}_2$  to the airflow promoted also the transport of ruthenium. It was concluded that the composition of gaseous atmosphere in the primary circuit has a significant effect on the amount of ruthenium transported to the containment during a severe accident.

Log: 390. **A NEW CONTINUOUS RADON MONITORING SYSTEM FOR CONTINUOUS MEASUREMENTS IN WATER.** Kil-Yong Lee; Soo Young Cho; Eunhee Lee; Sang-Ho Moon; Yoon Yeol Yoon; Dong-Chan Koh; Kyoochul Ha; Kyung-Seok Ko. Groundwater Department, Korea Institute of Geoscience and Mineral Resources (KIGAM) Gwahang-no 124, Yuseong-gu, Daejeon 305-350, Korea.

A new continuous radon monitoring (CRM) system was used for quantifying radon activity in natural water. The CRM system can measure radon in water continuously and discretely using an open or closed air loop. Since the RAD7 radon monitor that we used measure Po-218 as an indicator of Rn-222, one must wait for radioactive equilibrium between this parent-daughter pair. With a 3-min half-life, this only requires about 15 minutes. However, an additional equilibrium must be established between the radon-in-water and the radon-in-air. The CRM system was designed to speed up this process to allow higher resolution radon-in-water measurements. The design of the CRM system includes a nozzle as well as an impeller inside acryl tubing for rapid equilibration of radon between water and air phases. The design has also a shutoff valve in water discharge pipe to change the air-loop configurations. We compared the performance of the CRM to a traditional exchanger (RAD-AQUA, DurrIDGE) in air-water equilibration rates. The CRM system gave more rapid air-water equilibration using an open-loop but higher counts in a closed-loop configuration. As an example of how the device operates in the field, we show some in situ spatial mapping and time-series Rn-222 measurements for finding and quantifying submarine groundwater discharge (SGD) in Sogun Bay, South Korea.

Log: 391. **NEW THERMODYNAMIC DATA FOR TRIVALENT F-ELEMENTS AND HMBA COMPLEXES IN MIXED METHANOL WATER SOLVENTS.** Donley, A.S. (1); Clark, S.B. (2). (1) Washington State University. (2) Pacific Northwest National Laboratory.

Chemical separation of the trivalent lanthanides (Ln(III)) and actinides (An(III)) is commonly required to remove isobars or spectral interferences. Simple carboxylate ligands have been used extensively in chromatographic separations of the f-elements, yet some ion peaks remain difficult to resolve. Addition of a water-soluble co-solvent, such as methanol (MeOH), to the eluent can help achieve increased separation factors. The mechanisms that drive this phenomenon are poorly understood. In this study, the Ln(III)s were used as chemical analogs for the An(III)s due to their similar chemical behavior in solutions. A complete set of thermodynamic data in binary aqueous methanol solutions provides insight into the solvent effects and the forces that drive lanthanide-ligand complexation. Potentiometric, spectroscopic, and calorimetric titration experiments of Ln(III)s with 2-hydroxy-2-methylbutyric acid (HMBA) in 0, 10, 30, and 50% (vol%) methanol-water solutions revealed that the 1:1 and 1:2 complex formations become more exothermic with increasing MeOH fraction. There was no statistical change in the favorable entropy values with increasing MeOH concentrations. These results show that the Ln(III)-HMBA complexes are driven by both enthalpy and entropy, although the addition of methanol increases the equilibrium constants due to an increase in enthalpy and, by inference, improves the separation factors. As HMBA has a similar structure to that of alpha-hydroxyisobutyric acid (HIBA), it is expected that HIBA reactions are also enthalpy-driven in binary methanol water solvents. Further thermodynamic and separation experiments in different co-solvents, which disrupt the solvent structure of water, would test the hypothesis that enthalpy-driven complexation improves lanthanide separation.

Log: 392. **ANALYTICAL APPLICATIONS OF RADIOCHEMICAL EXTRACTANTS FOR CESIUM ISOTOPES.** DiPrete, D.P. (1), DiPrete C.C. (1). (1) Savannah River National Laboratory.

Waste cleanup at the Savannah River Site, as well as other DOE nuclear sites, has created an ongoing need to characterize the inventories of various waste tank heels prior to tank closure. Each tank heel has unique chemical and radiological distributions, rendering the use of routine analyses inadequate. Since each tank is a completely new matrix with often-unforeseen interferences, method development must be performed for each tank's residue material. Due to its high neutron-induced uranium fission yield coupled with a 30 year half-life, Cs-137 is one of the most predominant beta-emitting isotopes in DOE radioactive waste inventories. While Cs-137 is simple to quantify via the intense 662 keV gamma ray associated with the Cs-137/Ba-137m decay, the high abundance of this isotope in DOE waste streams can be problematic when quantifying other, significantly less abundant isotopes. The dose associated with Cs-137 is also problematic when large volumes of the matrix are required in order to meet sensitivity requirements. In order to reduce exposure and dose to laboratory personnel, as well as reduce analytical interferences and improve sensitivity, cesium removal protocols are often required

for radiochemical separation methodologies conducted on SRS high activity waste (i.e., tank heel) matrices. Ammonium molybdophosphate (AMP) is used extensively at SRNL as a Cs-removal agent in acidic solutions. AMP is useful to remove cesium from acidic matrices when it is an interfering radionuclide. However, situations occur in which cesium itself is the analyte of interest. An example would be the need to quantify Cs-135. While this isotope has a high neutron-induced uranium fission yield, it is much less radioactive than Cs-137 due to its long (2.3E6 year) half-life. Cs-135 is also a pure beta emitter, which makes analyses without very aggressive and successful separations from beta-emitting interferences, extremely challenging. Calixarene compounds, namely calix[4]arene-bis-(t-octylbenzo-crown-6), provide attractive alternatives to AMP for analytical applications requiring the recovery of a cesium isotope. Properties and applications of AMP and other specialized Cs-extractants for numerous radiochemical separation protocols for high activity residue will be reviewed.

Log: 393. **LOW ACTIVITY TEST SOURCES.** Porterfield, D.R. (1); Doyle, J.L. (1); Bland, G.J. (1); Yoho, M.D. (1 and 2). (1) Los Alamos National Laboratory. (2) The University of Texas at Austin.

We were confronted with the situation of needing to validate and test the function of new nuclear counting instrumentation (alpha spectrometry, gas-proportional, and liquid scintillation) in a new facility which has not yet been approved for radiological operations. The use of typical radioactive sources for this purpose required special approval and attendance by radiological protection personnel (RPP). To avoid this associated effort we looked at the use of sufficiently low activity sources that such approvals and attendance by RPP wouldn't be considered necessary. This poster will provide lessons learned on low activity test sources that we evaluated.

Log: 394. **Estimation of production date of uranium samples by determining  $^{230}\text{Th}/^{234}\text{U}$  isotopic ratios using MC-ICP-MS.** Song, K.(1);Lim, S.H.(1); Han, S.H.;Park, R.(1);Park, J.(1);Lee, C.-G.(1). (1) Korea Atomic Energy Research Institute. Radiochronometer techniques of uranium material have been being developed in many laboratories for nuclear forensic investigations. These techniques are based on the accurate ratio measurements of daughter-mother radionuclide pair such as  $^{230}\text{Th}/^{234}\text{U}$  and  $^{231}\text{Pa}/^{235}\text{U}$ . In particular,  $^{230}\text{Th}/^{234}\text{U}$  is frequently used isotope pair due to relatively rapid ingrowth of  $^{230}\text{Th}$  and availability of tracer and spike. For effective determination of accurate and precise  $^{230}\text{Th}/^{234}\text{U}$  ratio, either thorium isotopes have to be separated from relatively large amounts of uranium samples or extremely sensitive analytical methods have to be used. In both cases, rigorous chemical separation procedures are required to obtain pure Th isotopes prior to analysis. Additionally, the amounts of  $^{232}\text{Th}$  backgrounds during chemical procedures must be also minimized and characterized as far as possible in case of choosing  $^{232}\text{Th}$  as a spike. In this study, relatively simple and effective thorium separation method from uranium samples was adopted. Isotopic measurements of  $^{230}\text{Th}/^{234}\text{U}$  were then performed using highly sensitive MC-ICP-MS combined with isotope dilution mass spectrometry (IDMS). From these methodologies, over 90 % separation yields for  $^{230}\text{Th}$  from very small amounts of uranium samples (100  $\mu\text{g}$ ) have been achieved and uranium concentration after Th separation was below 0.1 ng (100 ppt). For IDMS, we added 500 ppt of  $^{232}\text{Th}$  spike to minimize the effect by  $^{232}\text{Th}$  backgrounds which were confirmed around 1~2 ppt in process blanks. The detection limit for  $^{230}\text{Th}$  quantification was identified as ~2 fg. In conclusion, we have tested radiochronometer techniques by using simple and effective chemical separation and highly sensitive MC-ICP-MS. We also performed accurate isotopic ratio measurement of  $^{230}\text{Th}/^{234}\text{U}$  and age-dating with uranyl nitrate samples.

Log: 395. **IMPROVEMENT OF ANALYTICAL PERFORMANCES FOR UTRA-TRACE AMOUNTS OF U AND PU IOSOTOPES BY USING MC-ICP-MS.** LIM, S.H.(1); HAN, S.-H.(1); PARK, R.(1); LEE, C.-G.(1); PARK, J.(1); SONG, K.(1) (1) KOREA ATOMIC ENERGY INSTITUTE.

Bulk analysis of environmental samples (ES) including U and Pu at ultra trace levels is important for monitoring undeclared nuclear activities for nuclear safeguards. For accurate and precise determination of quantities and isotopic ratios of U and Pu, minimizing uranium backgrounds in experimental environments, adoption of suitable chemical procedures for separating pure U and Pu isotopes and high sensitive measurement systems such as mass spectrometry are essentially necessary. For minimizing uranium backgrounds, we recently established a new clean facility called "CLASS (Clean Laboratory for Analysis of Safeguards Samples)" and Improvement in cleaning procedss for all labwares and strick managements of spikes and chemical reagents have been applied to reduce uranium backgrounds and prevent cross-contamination. We were able to confirm that uranium contents in process blanks were around 20 picograms levels through systemetic evaluations of uranium contents in process blanks during overal analytical procedures. We also improved chemical procedures including ashing, acid treatments and chemical separations in order to obtain higher chemical yields of U and Pu at ultra-trace levels in environmental samples. Quantification and isotopic measurements were then performed by using MC-ICP-MS (Neptune Plus, Thermo Scientific). More than 10 times enhanced signal sensitivity have been achieved by introducing and optimizing desolvation system (Aridus II, CETAC). Signal fluctuation in MC-ICP-MS have been reduced by adopting a peristaltic pump in the sample introduction system. As a part of verifications, analytical results with simulated samples including ultra-trace amounts of U and Pu isotopes will be presented along with such improved analytical procedures.



Log: 396. **Use of Boron and Strontium Isotopes for Groundwater Contamination Indicators by Agricultural Land Use.** Ko, K.S.(1); Ha, K.(1); Yum, B.W.(1); Koh, D.C.(1); Lee, K.Y.(1); Lee, K.K.(2). (1) Korea Institute of Geoscience & Mineral Resources (KIGAM). (2) Seoul National University.

This study were performed to know the factors controlling the groundwater chemistry such as agricultural land use by using  $\delta^{11}\text{B}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopes at the bank area of Mankyung River. Total 28 wells were used for the study and land use of the site was classified into two groups of rice paddy and greenhouse upland for Chinese cabbage and pumpkin cultivation. The results of shallow groundwater chemical analyses showed that the higher values of EC, Na, K, Ca, Cl, and  $\text{SO}_4$  were observed at upland area which was induced by the higher introduction of fertilizer and manure at upland than those of rice paddy area. On the other hand, the lower values of  $\text{NO}_3$  and redox potential of groundwater at paddy were lower than those at upland that were caused by the formation of reduced groundwater environment. The positive relationships between the isotopic ratios and concentrations of B and Sr indicated the strong effect of agricultural land use. PCA revealed that the first three factors explained around 72.2% of the total variance, which highlighted land use as the dominant factors affecting the groundwater quality. ANOVA results showed that there were significant differences ( $p>0.05$ ) of the parameters, including pH, EC, Na, Ca, Cl,  $\text{NO}_3$ ,  $\text{SO}_4$ , and B, between the two land use groups. From the study, the statistical methods of ANOVA and PCA could be used an effective method for demonstrating the relationship between land use and groundwater quality and to identify the main factors and sources of contamination.

Log: 397. **EXAMINATION OF LOCAL ATMOSPHERIC TRANSPORT OF RADIOXENON IN THE OTTAWA RIVER VALLEY.** Christine Johnson (1); Justin Lowrey (2); Derek Haas (2); Steven Biegalski (1). (1) The University of Texas at Austin. (2) Pacific Northwest National Laboratory.

Certain radioactive isotopes of the noble gas xenon are considered one of the primary indicators of a nuclear detonation. After an underground nuclear detonation certain radioxenon isotopes have an increased probability of detection and during an On-Site Inspection various methods are used in an effort to detect and quantify these isotopes. However, in order to effectively utilize these isotopes as indicators of nuclear testing an accurate understanding of other potential sources of radioxenon must be considered. A study was carried out in order to better understand the potential effects of local radioisotope production at a medical isotope production facility on an On-Site Inspection and to examine the transport mechanisms involved between release of radioxenon from a production facility and detection at a sampling location in the Ottawa River Valley in Ontario, Canada. Concentrations of the radioxenon isotopes Xe-133 and Xe-135 were measured as they were released from the stack at the Chalk River medical isotope production facility and were then measured at various sites in the Ottawa River Valley. Gaussian plume modeling was then used to model the transport of these radioxenon isotopes between the production facility and the sampling locations. Various corrections to the Gaussian plume equation are also examined for their impact on the local atmospheric transport of radioxenon.

**Log: 398. AN INTER-AGENCY APPROACH TO EXPLOITING FORENSIC EVIDENCE CONTAMINATED WITH RADIONUCLIDES: THE AUSTRALIAN EXPERIENCE.**

Toole, K.(1); Young, E. (1); Evans, T.(1); Roffey, P.(2); Chang, C.(2); Goodman-Jones, A.(2); Cho, K.(2); Shaw, T.(2); Burger, F.(2); Davies, B.(2); Reinhard, M.(1). (1) Australian Nuclear Science and Technology Organisation. (2) Australian Federal Police.

Research has demonstrated that some forensic evidence types (such as DNA, fingerprints and digital devices) may yield valuable information in spite of ionising radiation exposure, yet are not amenable to decontamination. Thus, the development and validation of capabilities for the safe handling and exploitation of forensic evidence contaminated with radionuclides is an important step for a State in preparing to respond to and investigate a nuclear security event. In Australia, this capability is shared by the Australian Nuclear Science and Technology Organisation's (ANSTO) Nuclear Forensic Research Facility (NFRF) and the Australian Federal Police (AFP), with AFP forensic scientists undertaking examination of evidence in the NFRF. NFRF staff, who have experience in both forensic science and nuclear science, form a key part of the analytical team by serving as the 'interface' between these disciplines. The focus of this capability has been evidence types and examination techniques most likely to yield operational intelligence and/or highly probative evidence for court proceedings. Facilities and techniques have been amended or developed where required to suit the constraints inherent to radioactive material handling whilst still meeting the requirements of law enforcement, such as sample preservation, avoidance of cross-contamination and maintenance of chain of custody. Additionally, operationally-driven research is being undertaken to validate existing forensic procedures for; DNA extraction (utilising commercially available kits); fingerprint development and enhancement and; digital evidence collection on samples contaminated with radionuclides. The nuclear forensic capabilities being implemented by ANSTO and the AFP are not widely available in the Asia-Pacific region.

Log: 399. **Study on sorption of Cs, Sr and Co for Bentonite-Rock Mixtures under various ionic strength effects.** Yu-Lin Tu(1);Chuan-Pin Lee(1);Ming-Chee Wu(1);Shih-Chin Tsai(2);Ching-Yuan Liu(3);Tsuey-Lin Tsai(4).(1)Department of Earth Sciences, National Cheng Kung University, Tainan 70101, Taiwan.(2) Nuclear Science and Technology Development Center, National Tsing Hua University, Hsinchu 30013, Taiwan.(3)Department of Chemical and Materials Engineering, National Central University, Taoyuan 32001, Taiwan. (4)Chemistry Divisions, Institute of Nuclear Energy Research, Taoyuan 32546, Taiwan.

The buffer/backfill materials for radioactive waste disposal repository site were made of pure bentonite or bentonite-rock mixtures. This study was subjected to obtain the sorption capability of buffer/backfill materials to the important radionuclides such as Cs, Sr and Co by batch method for mixture of the host rock (argillite and granite) with either the U.S. Wyoming MX-80 bentonite or the Taiwan domestic Zhi-Shin bentonite in various proportion (0 ~ 100%). According to the experimental results, the distribution coefficients (Kd) of Cs, Sr, and Co obtained for the bentonite-rock mixtures had shown an approximate value to that of pure bentonite; for mixtures to have more than 50% of mixing proportion for bentonite to argillite or granite. Furthermore, it was obviously found that sorption to Cs Sr and Co for bentonite-rock mixtures decreases as ionic strength increased from 0.001 to 1M in NaCl solutions. According to the experimental process, in synthetic groundwater, it would be quite convenient and helpful to assess the distribution coefficients (Kd) of Cs, Sr, and Co to the buffer/backfill materials by batch sorption experiments with bentonite-rock mixtures of fixed proportion.

Log: 400. **HALF-LIFE MEASUREMENTS OF BERYLLIUM-7 IN MATERIALS.** Ohtsuki, T.

The formation of atom-doped C60 and C70 etc. has been investigated by using several types of radionuclides produced by nuclear reactions. From the trace of the radioactivities after high performance liquid chromatography (HPLC), it was found that formation of endohedral fullerenes of Be atom is possible by a recoil process following the nuclear reaction. The decay rate of 7Be electron capture (EC) was measured in C70 and Be metal with a reference method. The half-lives of 7Be endohedral C70 and 7Be in Be metal (Be metal(7Be)) were found to be  $52.45 \pm 0.04$  and  $53.25 \pm 0.04$  days, respectively. This amounts to a 1.5% difference in the EC-decay half-life between 7Be@C70 and Be metal(7Be). The results are a reflection of the different electron wave-functions in nuclear site for 7Be inside C70 compared to when 7Be is in a Be metal. The further theoretical interpretation is needed for these experimental results.

**Log: 401. PROTACTINIUM-231 MEASUREMENT FOR ISOTOPE CHRONOMETRY IN NUCLEAR FORENSICS.** Keegan, E; Stopic, A; Griffiths, G. ANSTO.

Using two separate parent/daughter isotopic pairs to date unknown nuclear material as part of a nuclear forensic investigation provides increased confidence in the material's analytically measured age. For this reason, the Australian Nuclear Science and Technology Organisation (ANSTO) is currently developing the capability to measure Pa-231 (half-life = 32,760 years) by mass spectrometry to enable application of the Pa-231/U-235 chronometric system for uranium bearing samples in addition to the more commonly applied Th-230/U-234 system. As part of this development, a review of the documented radiochemical separation procedures of Pa from Th and U, using both extraction chromatography and anion exchange techniques, was undertaken. Radiochemical separation experiments were carried out to verify and expand on existing literature data, and determine the optimal separation procedure. For these experiments we used the beta/gamma emitting nuclide Pa-233 (half-life = 26.97 days) produced by neutron irradiation of Th-232 in ANSTO's OPAL research reactor. Measurements of Pa-233 were performed using gamma spectroscopy (for initial activity) and gamma counting (for separation efficiency and chemical yield determination). The results of these experiments will be described.

**Log: 402. LASER ABLATION OF SIMULATED USED NUCLEAR FUELS.** Campbell, K. (1); Unger, A. (1); Bertoia, J. (1); Hartmann, T. (1); Czerwinski K. (1). (1) University of Nevada Las Vegas.

The ability to directly examine elemental and isotopic concentrations in fresh fuels, used fuels, and actinide containing material without dissolution of the solid phase is of interest to Nuclear Safeguard and Forensic programs. The application of laser ablation-inductively coupled plasma mass spectrometry (LA-ICP-MS) to determine isotope ratio and elemental measurements respectively, grants superior technology for direct solid sampling in analytical chemistry. The advantages of these techniques include reduced risk of contamination by direct sample analysis, no chemical dissolution, and determination of spatial distributions of elemental compositions. This work focuses on uranium oxide simulated used nuclear fuels starting with binary systems of (U,Pu)O<sub>2</sub>, (U,Np)O<sub>2</sub>, (U,Ce)O<sub>2</sub> and (U,Zr)O<sub>2</sub>. Methodology was successful in observing linearity of 0.995 and greater for these systems. This was achieved by minimizing the particle size distribution of the aerosol and in turn decreases the time-dependent fractionation often observed in LA-ICP-MS.

Log: 403. **COMPLEXATION AND EXTRACTION STUDIES OF HIGH VALENCY ACTINIDES BY SCHIFF BASE LIGANDS.** Bustillos, C.G.(1,2); Hawkins, C.A.(1); Copping, R.(2); May, I.(2); Nilsson, M.(2). (1) University of California Irvine. (2) Los Alamos National Laboratory.

Increased knowledge of actinide coordination chemistry and the development of advanced actinide separation processes are essential to reducing the radiotoxicity of used nuclear fuel. Commercial separation techniques for nuclear fuel have focused on the selective extraction of U(VI) and Pu(IV), while contemporary efforts are aimed at extracting the minor actinides, e.g. Np and Am. These four mid-actinides (U, Np, Pu, Am) have accessible higher oxidation states (+V, +VI) at which they exist as linear dioxo actinyl ions  $[AnO_2]^{n+}$ , offering the possibility of co-extraction, or back-extraction of all 4 actinide cations. The  $An=O$  bonds permit ligand coordination in the equatorial plane of the metal ion center, affording a geometry for unique bonding characteristics. Schiff bases can be prepared with synthetic ease and offer essentially planar chelation around the equatorial plane of actinyl ions through their N(2)O(2) binding site, presenting the possibility that this distinct coordination environment may facilitate the selective solvent extraction or aqueous retention of pentavalent and hexavalent actinides. Preliminary results indicate that these ligands show promise for group solvent extraction or retention of the mid-actinides. Lipophilic salen-based ligands prepared in our labs are able to extract uranium from nitrate media with 90% or higher extraction in one contact. These ligands have been shown to form 1:1 complexes with U(VI), Np(VI) and Pu(VI) which have been structurally and spectroscopically characterized. Such ligands show promise for group solvent extraction or retention of the mid-actinides and recent results will be presented.

Log: 404. **RADIONUCLIDES IN THE NEW ZEALAND DIET.** Golovko, O.(3); Pearson, A.(1); Gaw, S.(2); Hermanspahn, N. (3); Glover C.(2). (1) Ministry for Primary Industries, Wellington, New Zealand (2) University of Canterbury, Christchurch, New Zealand (3) Institute for Environmental Science & Research, Christchurch, New Zealand.

As a result of the accident at Fukushima-Daiichi NPP concerns have been raised in New Zealand over anthropogenic radionuclides entering into the diet, particularly from consumed seafood. New Zealand has historically monitored milk as a sentinel of dietary levels of fallout radionuclides, the appropriateness of this as a monitoring method post-Fukushima however is unknown. In addition there is only a limited record on NORM levels within the diet. To address this research has been undertaken to derive activities across a variety of food types for important anthropogenic and natural radionuclides, from which baselines for the New Zealand diet have been established. Caesium137 was detected in 8.75% of samples, based on results a level of 0.05 Bq/kg is the estimated baseline for terrestrial species and up to 0.5 Bq/kg for marine species. Polonium210 was present in the majority of foods, with maximum activities of 20-30 Bq/kg detected in shellfish. Uranium isotopes were also present in a number of foods. The collected results have been used to refine the estimated contribution of dietary radionuclides to the total population exposure to ionising radiation. Complementary to the dietary baseline survey a detailed analysis of Caesium137 and Polonium210 activities in a range of seafood species from six New Zealand fishing regions has been conducted. This is to inform future monitoring programs for seafood as well as provide insight into exposures for potentially at high risk groups, such as seafood subsistence consumers.

Log: 405. **TRACE ELEMENT MICRO DISTRIBUTION IN NAIL CLIPPINGS.** Favaro, P.C.(1); Bode, P.(1); De Nadai Fernandes, E.A. (2). (1) Delft University of Technology (2) CENA, University of Sao Paulo.

Sometimes extensive cleaning procedures are needed prior to analysis of nail clippings. These procedures can vary from mild washing to mechanical scraping. Water, detergents, acetone, alcohol and acids are used in washing procedures. Though the procedures aim at removal of the exogenous elements, it cannot be excluded that they also may result in leaching of the endogenous chemical elements. The scraping process removes part of the sample and depending on the chemical distribution of the chemical elements in the nail keratin, with the possibility of concentration or dilution of the chemical element in the sample. Understanding the micro distribution and mobility of the chemical elements through the nail clipping is import to identify and avoid errors during the pre-treatment of the samples. Micro-beam PIXE (Proton Induced X-Ray Emission) has been used for measurement of the element distributions along the thickness of the nail. It allowed for assessing if the cleaning procedures results in the leaching of endogenous chemical elements, and also if the scraping process leads to disproportional removal of these elements.

Log: 406. **Development of an Ultra-High Intensity Pulsed Neutron source using a Pulsed Laser Source.** George H Miley University of Illinois.

We are developing a high intensity pulsed neutron source for homeland security applications and also various NAA uses. The key feature involves the use of deuteron clusters created in a thin palladium foil. The deuterons are driven out of the foil at multi-MeV energy by irradiation with a pulsed Petawatt laser. This technique has been demonstrated in experiments using the TRIDENT pulsed Petawatt laser at LANL. The neutron source uses the intense MeV deuteron beam to bombard a plate (or "target") containing deuterons or tritium. D-D reactions in the former yield 2.45-MeV neutrons, or with D-T reactions would provide 14.7-MeV neutrons. A high repetition rate is achievable using a target interaction chamber with targets injected rapidly as envisioned for Inertial Confinement Fusion. This design automatically handles the heat load issue encountered in other designs. This facility can generate deuteron beam intensities much higher than any of common accelerator – target designs. In addition, the use of multi MeV deuteron ions, near the energy of the fusion reaction neutrons (versus keV ions in accelerator type experiments) result in a distinct forward direction for the neutron reaction. The estimated deuteron yield  $>10^{15}$  neutron/pulse for D-T neutrons, or  $>10^{13}$  neutron/pulse for D-D neutrons assuming a 100-kJ laser with 10% conversion efficiency. A larger laser with higher conversion efficiency will give a much larger deuteron yield and higher deuteron energy. This adds to the efficiency by reducing neutron losses in the undesired directions and reducing chamber shielding requirements.

Log: 407. **PREPARATIVE SEPARATION AND PURIFICATION OF THULIUM FROM ERBIUM FOR NEUTRON CAPTURE CROSS SECTION MEASUREMENTS.** Bene, B. J.(1); Taylor, W. A.(2); Birnbaum, E. R.(2); Sudowe R.(1). (1) University of Nevada, Las Vegas. (2) Los Alamos National Laboratory.

The determination of the low-energy neutron capture cross-section of Tm-171 with greater accuracy is important for both astrophysics and Stockpile Stewardship. Thulium-171 is a branching point in the s-process of stellar nucleosynthesis. Branching point nuclei, with half-lives in the order of 1 – 100 years are places where neutron capture and beta decay competes. This isotope is also the second order (n,  $\gamma$ ) reaction product of Thulium-169, which was used as a radiochemical detector that was placed at strategic locations in nuclear devices during the era of weapon testing. Measurement of the neutron capture cross-section of Tm-171 will provide better insight into physical processes in stars as well as nuclear devices. Thulium-171 is planned to be produced through a neutron capture reaction on isotopically enriched stable Er-170 followed by beta decay. After irradiation, milligram amounts of thulium will need to be separated from the remaining target material of about half a gram. A highly efficient and very selective separation based on ion exchange chromatography has been developed to separate these neighboring lanthanides in preparative quantities. The optimized single pass-through separation procedure, its reproducibility and a comparison of the Eichrom RE2 and branched-DGA resins for the purification of the thulium fraction will be presented.

Log: 408. **ACTIVATION ANALYSIS IN GEOCHEMICAL CHARACTERIZATION OF AUSTRALASIAN TEKTITES AND LOCALIZATION OF THEIR POSSIBLE PARENT CRATER.** Mizera, J.(1,2); Řanda, Z.(1); Kameník, J.(1). (1) Nuclear Physics Institute ASCR, Czech Republic. (2) Institute of Rock Structure and Mechanics ASCR, Czech Republic.

Australasian tektites (AAT) of various morphological types (splash-form, ablated splash-form, layered - Muong Nong) from various sites in Southeast Asia, South China and Australia were geochemically characterized by methods of instrumental neutron and photon activation analyses. Based on the acquired data as well as previously published geochemical, mineralogical, petrographic and isotopic data on both AAT and their potential source materials, and considering ballistic, palaeogeographic, palaeoclimatic and palaeoecological constraints known from other tektites and their strewn fields, probable source materials and location of the impact site and parent crater for AAT are discussed. Criticism has been raised against previously suggested localization of the impact in Indochina and supporting evidence has been brought for revisiting Chinese loess or its precursors as the AAT parent materials. Localization of the parent crater in the deserts of North Central China has been suggested. The study was supported by the grant 13-22351S of the Czech Science Foundation.

Log: 409. **EXPERIMENTAL STUDY ON SIZE DISTRIBUTION OF FISSION-PRODUCT AEROSOLS.** Takamiya, K.(1); Tanaka, T.(1); Nitta, S.(1); Itosu, S.(1); Sekimoto, S.(1); Oki, Y.(1); Ohtsuki, T.(1). (1) KURRI.

A part of fission products released in the air from FDNP have been transported as aerosols. Kaneyasu et al. reported sulfate was the potential transport medium for radioactive cesium because of the coincidence of the activity size distributions of radiocesium and the mass size distribution for non-sea-salt sulfate. However, mechanism of adsorption of fission products to sulfate aerosol has not been clear. In the present work, adsorption behavior of fission products to various aerosol has been observed to elucidate the adsorption mechanism. Aerosols were generated by using a combined system of an atomizer, heater and diffusion dryer. The atomizer generates aerosol particles from a solution. Sodium chloride and ammonium sulfate solutions of various concentrations were used in the present experiments. The generated sodium chloride or ammonium sulfate aerosol particles were passed through an electric furnace or a fission chamber to attach radioisotopes on aerosols to produce radioactive aerosols. In the furnace, radioisotopes such as radiocesium compound were heated to vaporize. And Cm-248 emitted fission fragments in the fission chamber. The radioactive aerosols were injected to a cascade impactor to classify aerosol particles according to their sizes. Gamma-ray spectroscopy was performed to the size-classified radioactive aerosols. Finally measured activity size distributions and the mass size distributions were compared to obtain the size dependence of adsorption of radioisotopes.



Log: 410. **NUCLEAR WASTE GLASS ALTERATION UNDER GAMMA IRRADIATION IN PRESENCE OF NEAR-FIELD MATERIALS.** Parruzot, B.(1); Waters, S.M.(2); Wall, D.E.(2); Neeway, J.J.(3); Ryan, J.V.(3); Qafoku, N.P.(3); Gin, S.(4); Wall, N.A.(1)\*. (1) Washington State University, Chemistry Department, Pullman, WA, USA. (2) Washington State University, Nuclear Radiation Center, Pullman, WA, USA. (3) Pacific Northwest National Laboratory, Energy and Environment Directorate, Richland, WA, USA. (4) CEA Marcoule, DEN/DTCD/SECM, Bagnols sur Cèze, France. \*Corresponding author.

In several countries, borosilicate glass is considered or used as the confinement matrix for high level nuclear waste – fission products and minor actinides. The glass canisters are to be stored in deep geological repositories, where long-term safety must be demonstrated. Ground water and near-field materials may impact glass alteration mechanisms and rates in repository conditions. Iron and iron corrosion products originating from the stainless steel glass canister degradation are known to enhance the precipitation of specific ferrosilicate phases outside and within the gel layer formed at the glass surface during its alteration. Under irradiation, radiolysis of the solution may lead to specific redox conditions impacting iron mobility and the nature of the precipitated phases. To assess this impact, International Simple Glass (ISG, a 6-oxide simple glass used as international benchmark) and SON68 glass (non-radioactive analogue of the French HLW waste glass) were altered under gamma irradiation (Co-60 source, 270 Gy/h) at 90°C in the presence of iron or hematite ( $\text{Fe}_2\text{O}_3$ ). Experiments were carried out under argon atmosphere to maintain a nitrogen-free environment, thus avoiding the formation of nitric acid upon radiolysis. This work will present short-term glass alteration data, focusing on solution analysis and characterization of the iron oxidation state evolution under irradiation. This work will be a base for future experiments aiming to better characterize the long-term evolution of such systems. This work is supported by DOE/NEUP, under Project 23-3361.

Log: 411. **CORROSION OF THE LOW-ACTIVITY CEMENTIOUS WASTE FORMS SALTSTONE AND CAST STONE BY NUCLEAR WASTE REPOSITORY GROUNDWATERS.** Reiser, J.T.(1); Riggan, B.K.(1); Musa C.M.(1); Parruzot, B.(1); Neeway, J.J.(2); Ryan, J.V.(2); Qafoku, N.P.(2); Wall, N.A.\*(1). (1) Washington State University, Chemistry Department, Pullman, WA, USA. (2) Pacific Northwest National Laboratory, Energy and Environment Directorate, Richland, WA, USA. \*Corresponding author.

Cementitious low-level nuclear waste forms are produced at DOE cleanup sites (e.g. Savannah River) for the confinement of long-lived radioelements. The leachability of two non-radioactive representative cementitious materials, Cast Stone and Saltstone, was tested at 90°C for 30 days at a surface-area-to-solution-volume ratio of 2000 m<sup>-1</sup> in deionized water as well as in nine repository-relevant groundwaters (Brine, Lime, Boom clay, Grimsel, Yucca mountain, Savannah River, Hanford, WIPP GWB and WIPP ERDA-6). Preliminary data indicate a fast pH(90°C) increase to values over 10 after one hour of alteration followed by a plateau up to 30 days. pH in highly concentrated ground waters is constant (Yucca mountain and ERDA-6) or decreasing (GWB). Results for solution elemental analyses and solid characterizations (SEM-EDS and powder XRD) will be presented to show the phenomenology of the Cast stone/Saltstone alteration (release of elements from the waste form, formation of secondary phases.). This work is supported by DOE/NEUP, under Project 23-3361.

Log: 412. **DETERMINATION OF MINERAL SURFACE AREA BY METHYLENE BLUE SORPTION: COMPARISON WITH BET AND GEOMETRIC SURFACE AREAS.** Parruzot, B.(1); Reiser, J.T.(1); Riggan, B.K.(1); Wall, N.A.(1)\*. (1) Washington State University, Chemistry Department, Pullman, WA, USA. \*Corresponding author.

In order to accurately quantify mineral weathering rates in the context of nuclear repository performance, mineral reactive surface areas must be determined with precision. A calculated geometric surface area may be used as a first approximation, but BET gas adsorption technique is commonly used and takes into account the effective surface area and, if existing, cracking of the mineral grains. However, constraints during BET measurement such as use of vacuum and high temperatures are susceptible to affect the mineral structure and thus induce bias in the measured surface area value [1]. In this study, surface areas of iron corrosion products and glasses of interest for nuclear waste management are measured via methylene blue sorption using UV-Visible spectrophotometry and by BET. The results from both techniques were compared to calculated geometric surface area and the applicability of each technique to components relevant to high-level nuclear waste repositories are determined. This work is supported by DOE/NEUP, under Project 23-3361. [1] Wall N.A. et al. Chem Geol v. 274, 149-157 (2010)

Log: 413. **Daily intakes of 40K and 137Cs in mainly consumed foods in Korea.** Chae, J.S.(1); Song, M.H.(1); Lee, J.H(1); Yun, J.Y(1). (1) Korea Institute of Nuclear Safety.

Humans are exposed to several kinds of external and internal radiation sources such as terrestrial radiation, cosmic radiation, inhalation and ingestion of natural and artificial radionuclides. Ingestion of food containing radionuclides contributes to substantial portion of effective internal dose. 40K is one of the most important natural radionuclides because potassium is an essential element of cellular tissue. Over 50% of effective internal dose through daily dietary intake is originated from radioactive potassium. 137Cs is an artificial radionuclide with long half-life of about 30 years. It can be released to the environment from nuclear industry and nuclear accidents and eventually, entered the human bodies after transporting along various environmental pathways. In order to estimate radiological effects due to ingestion of food, activity concentrations of 40K and 137Cs in mainly consumed foodstuffs in Korea were evaluated. Moreover, the amount of intake of these radionuclides and annual internal effective dose resulting from ingestion of radionuclides were calculated.

Log: 414. **Comparison and optimization of digestion methods for uranium analysis in soil by alpha-spectrometry.** Chae, J.S.(1); Kim, C.J.(2). (1) Korea Institute of Nuclear Safety. (2) Korea Atomic Energy Research Institute.

The measurements of concentration of uranium isotopes in various environmental samples are very important in many fields. The alpha spectrometry is one of the most common methods for uranium isotopes analysis because of its high sensitivity and low cost. Sample decomposition is necessary before chemical separation step using extraction chromatography. Various methods are used for acid leaching or digestion with hot plate or conventional microwave digester for solid samples, and fusion is an appropriate digestion method for reducing hazardous acid and emission of acid vapors. In this study, the precision and accuracy of various decomposition methods for analysis of uranium isotopes using alpha spectrometry with extraction chromatography are investigated. In addition, the appropriate condition for sample decomposition such as acid type, acid amount and duration time of sample attack are estimated.

Log: 415. **STRUCTURAL INVESTIGATIONS OF METAL-ION COMPLEXATION AND AGGREGATE FORMATION COMBINING TBP AND HDBP: INSIGHTS FROM X-RAY ABSORPTION SPECTROSCOPY.** Braatz, A.D.(1); Ellis, R.(2); Antonio, M.(2); Nilsson, M.(1). (1) Department of Chemical Engineering and Materials Science, University of California, Irvine. (2) Chemical Sciences and Engineering Division, Argonne National Laboratory.

Advanced nuclear fuel cycles are dependent on successful chemical separation of the various elements present in the used fuel. Numerous extraction systems have been developed for the recovery and separation of the various metal ions present in the used fuel. In some of these solvent extraction systems, aggregation phenomena can interfere with the efficiencies of the processes and decrease the successfulness of the separation. To investigate the aggregation more closely, 1 M combined solutions of tri-n-butyl phosphate (TBP) and dibutyl phosphoric acid (HDBP) in n-dodecane were chosen as the organic phase to be contacted with an aqueous phase containing a single metal ion from the lanthanide series in either 2 M or 0.2 M nitric acid. The extraction of lanthanum, dysprosium and lutetium from nitrate solutions at various metal ion concentrations was investigated. Distribution ratios of the metal ions between phases will be presented combined with water uptake to help determine the physicochemical properties of the investigated system. X-ray absorption behavior of these organic solutions will be presented to shed light on the macromolecular structures created by the extractants. Possible structures and possible explanations of the physicochemical properties will be presented.

Log: 416. **DETERMINATION OF ISOTOPIC RATIOS OF PLUTONIUM AND URANIUM IN SOIL SAMPLES BY THERMAL IONIZATION MASS SPECTROMETRY.** Shibahara, Y(1); Kubota, T(1); Fujii, T(1); Fukutani, S(1); Takamiya, K(1); Konno, M(2); Mizuno, S(2); Yamana, H(1). (1) Kyoto University Research Reactor Institute, (2) Fukushima prefectural government.

Isotopic ratios of plutonium and uranium in soil samples were investigated, in order to understand the level of contamination with plutonium and uranium released at the accident of Fukushima Daiichi nuclear power plant (FDNPP). As references, NIST (SRM4354, SRM4357) and IAEA (IAEA368, IAEA384) standard reference materials (soils), which are contaminated by high active waste from reprocessing plant or fallout from nuclear test, were chosen. Plutonium and uranium were recovered from the reference samples by extraction/ion-exchange combination chromatography and their isotope compositions were analyzed by thermal ionization mass spectrometry. The isotopic ratios of plutonium (Pu-240/Pu-239) and uranium (U-235/U-238) agreed well with reported data analyzed by ICP-MS. By applying our analytical technique to environmental soil samples obtained in Fukushima prefecture, a possibility of atmospheric release of nuclear fuel matrix during the accident of FDNPP is discussed.

Log: 417. **PNEUMATIC TUBE DELIVERY SYSTEM FOR SHORT-TIME IPAA AT THE MT-25 MICROTRON.** Horák, Z.(1); Chvátil, D.(1); Krist, P.(1); Olšanský, V.(1); Krausová, I.(1); Mizera, J.(1,2); Řanda, Z.(1). (1) Nuclear Physics Institute ASCR, Czech Republic. (2) Institute of Rock Structure and Mechanics ASCR, Czech Republic. Microtron MT-25 of the Nuclear Physics Institute ASCR is an effective source of high energy photon radiation utilized in the instrumental photon activation analysis (IPAA). Recent modernization of MT-25 included installation of a pneumatic tube delivery system (PTDS) designed for fast transport of samples between the irradiation position and an HPGe detector. PTDS is driven by three vacuum cleaners. A rotating switch placed above the detector shielding switches between four routes for (i) manual sample insertion, (ii) sample delivery to the irradiation position and back, (iii) sample delivery to the counting position, and (iv) discarding sample. A vertical rotating irradiation terminal defines sample position during irradiation. PE vials of 25 ml volume are used as irradiation capsules. Transport and switching times have been minimized so that counting can be started 6 seconds after the end of irradiation. Both the spectrometer and PTDS are controlled from a single PC. The PTDS control program allows automated operation with preset irradiation, decay and counting times, with possibility to manipulate with sample between irradiation and counting (e.g., to repack sample for lower blank). Cyclic irradiation is also possible. Examples of recent applications of PTDS in the short-time IPAA are presented. The work has been supported by the project 13-27885S of the Czech Science Foundation.

Log: 418. **MEASUREMENTS OF THE NEUTRON ACTIVATION CROSS SECTIONS FOR BI AND CO AT 134 MEV.** Yashima, H. (1); Sekimoto, S. (1); Ninomiya, K. (2); Kasamatsu, Y. (2); Shima, T. (2); Takahashi, N. (2); Shinohara, A. (2); Matsumura, H. (3); Satoh, D. (4); Iwamoto, Y. (4); Hagiwara, M. (3); Nishiizumi, K. (5); Caffee, M. W. (6); Shibata, S.(7) (1) Kyoto University (2) Osaka University (3) KEK (4) JAEA (5) University of California (6) Purdue University (7) RIKEN.

Activation cross sections, by high-energy neutrons, are used by cosmochemists to decipher the cosmic-ray irradiation histories of meteorites. These cross sections are also used to estimate residual radioactivities at accelerator facilities. While there is ample data for the neutron production of radionuclides below 20 MeV, neutron activation cross sections above 20 MeV are scarce. We are measuring high-energy neutron activation cross sections of Bi and Co using a quasi-monoenergetic neutron beam. The quasi-monoenergetic neutrons are produced using the Li-7(p,n) reaction at the ring-cyclotron of Research Center for Nuclear Physics (RCNP), Osaka University. Since the energy spectrum of this neutron field through the Li-7(p,n) reaction has a high energy peak (134 MeV) and a low energy tail, two neutron beams for 0 and 25 degree for proton beam axis were used for sample irradiation to correct for these low energy tails. The neutron energy spectra were measured by using liquid organic scintillator and time-of-flight (TOF) technique. After irradiation, gamma-rays were measured with a HPGe detector. The reaction rates were obtained from gamma-ray spectrum. The neutron activation cross sections were estimated by subtracting the reaction rates of samples for the 25 degree irradiation from those of the 0 degree irradiation. In this conference, we will present these experimental cross section data, combined with other experimental data (neutron-induced and proton-induced), and calculations using the PHITS code.

Log: 419. **STUDY OF ORGANIC MATTER SOURCES USING  $\Delta^{14}\text{C}$ ,  $\delta^{13}\text{C}$  AND PLUTONIUM ISOTOPE SIGNATURES.** Lujanienė, G. (1); Mažeika, J. (2); Li, H.-C. (3); Petrošius, R. (1, 2); Barisevičiūtė, R. (1); Jokšas, K. (2); Remeikaitė-Nikienė, N. (1,4); Malejevas, V. (1,4); Garnaga, G. (4); Stankevičius, A. (1,4); Kulakauskaitė, I. (1); Šemčuk, S. (1); Povinec, P.P. (5). (1) SRI Center for Physical Sciences and Technology, Lithuania. (2) SRI Nature Research Centre, Lithuania. (3) NTUAMS Laboratory, National Taiwan University. (4) EPA, Lithuania. (5) Comenius University, Bratislava, Slovakia.

Pu isotope ratios in bottom sediments (BSs),  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$  of total organic carbon (TOC), as well as in humic acids (HAs), lipid and phospholipids (PL) fractions of the BSs were applied to study sources and fate of organic carbon (OC) in the Baltic Sea including dumpsites of chemical warfare agents (CWA). The compound-specific  $\delta^{13}\text{C}$  analysis, PL-derived fatty acid biomarkers and a different end-member mixing model approach were used to estimate the relative contribution of the marine, terrestrial and fossil sources to OC in the coastal and open sea BSs, to assess a possible effect of the petroleum hydrocarbon (HC) contamination on radiocarbon signatures and to elucidate a probable leakage of CWA at the CWA dumpsite in the Gotland Deep. The obtained data revealed the possible application of the Chernobyl-derived Pu to trace the pollutants of the terrestrial origin. Wide OC variations with the strong impact of the terrestrial and fresh waters in the coastal areas were observed. It has been shown that similar values of  $\delta^{13}\text{C}$  found in HA and TOC are due to the high abundance of HAs in the sediments. Variations of  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$  values in total lipid fractions (TLF) have indicated the mixed sources of lipids in the coastal area. The most depleted values were detected for the  $\Delta^{14}\text{C}$  of TOC ( $-453\text{‰}$ ) and TLF ( $-812.4\text{‰}$ ) at the CWA dumpsite. The contribution of HC to the fossil fractions was estimated from measurements of HC in BSs. On the average, HC, terrestrial, marine and fossil sources accounted for 1%, 32%, 50%, and 17% of OC in BSs, respectively. A high contribution of fossil sources at the CWA dumpsite as compared to other stations in the Baltic Sea was found. This study was supported by the Research Council of Lithuania (contract No. MIP-080/2012).

**Log: 420. SORPTION OF RADIONUCLIDES AND HEAVY METALS TO NATURAL CLAY, GRAPHENE OXIDE AND GRAPHENE OXIDE-FERROUS OXIDE COMPOSITE.**

Lujanienė, G. (1); Kulakauskaitė, I. (1); Šemčuk, S. (1); Tautkus, S. (2); Valiulis, D. (1); Mažeika, K. (1); Motiejūnas, S. (3). (1) SRI Center for Physical Sciences and Technology, Vilnius, Lithuania. (2) VU Faculty of Chemistry, Vilnius, Lithuania. (3) SE Radioactive Waste Management 〇敲据<sup>9</sup>〇〇嘜 w 魷瑩〇〇〇〇 A Waste manage.

Waste management and related problems are among the most relevant ones in Lithuania and around the world. Technologies based on the natural locally available clay or other minerals are the most widely used for engineered barriers due to their cost-effectiveness and high reliability. Currently graphene oxide and graphene composites are supposed to be new promising materials for the next generation barriers in the environment protection. The adsorption capacities of natural clay, graphene oxide and graphene oxide-ferrous oxide composite were compared with the aim of their possible application in the environmental protection technologies. Graphene oxide and graphene composites were synthesized using methods described in literature and characterized by SEM, XRD FTIR and Mössbauer spectroscopy. Sorption of Am and Pu isotopes as well as Co, Ni, Cu and Pb to natural clay, graphene oxide and graphene oxide-ferrous oxide composite was studied in equilibrium and kinetic experiments. Concentrations of elements in the liquid-phase were measured using ICP-MS and alpha – spectrometry. Studies of sorption pH dependences showed wide variations in the uptake of studied elements.

**Log: 421. Updated meteorological analysis on a possible nuclear test in DPRK in May 2010.** Wotawa, G. (1); Maurer, C. (1) (1) Zentralanstalt fuer Meteorologie und Geodynamik.

Between 13 and 22 May 2010, multiple detections of radionuclides were recorded at CTBTO monitoring stations located in Eastern Asia. Other studies indicated additional radioxenon detections on 13 May 2010 at a monitoring station located on the territory of the Republic of Korea. A comprehensive atmospheric transport analysis presented at the MARC-IX conference showed that the fission products could have been released from locations on the territory of DPRK. Other release areas, however, could not be ruled out. Recent studies showed evidence of a seismic event on 12 May 2010 at the facility where the announced nuclear tests of 2006, 2009 and 2013 were performed. Based on the new results, the study presented at the last MARC conference is updated, and the consistency between the seismic event and the radionuclide events is shown.



Log: 422. **Simulation of transport and deposition of radionuclides from the Fukushima Daiichi nuclear accident.** Wotawa, G. (1), Arnold, D. (1), Maurer, C. (1) (1) Zentralanstalt fuer Meteorologie und Geodynamik.

In a study performed as part of the WMO Technical Team on the Fukushima accident, the role of increasing horizontal resolution and improving precipitation input data on FLEXPART modelling results was investigated. It was found that increasing meteorological resolution of the ECMWF input analyses and ingesting precipitation data from the Japanese MESO analyses lead to good overall deposition modelling results, compared with the monitoring data. The direct ingestion of radar and rain gauge analyses did not improve the model predictions. It was also shown that existing deposition parameterization schemes may require some modifications if better resolved meteorological input data are used. On the impact side, the Fukushima case was used to investigate how comparable nuclear accidents may impact air traffic and aviation, and what immediate measures could be taken in the event of an emergency.

Log: 423. **OPTIMIZING DETECTOR PARAMETERS FOR IN SITU LOW ENERGY GAMMA SPECTROMETRY IN CLOSE GEOMETRIES.** Hedman, A.(1,2); Bahar Gogani, J.(1); Ramebäck, H.(1,3). (1) Swedish Defence Research Agency. (2) Umeå University. (3) Chalmers University of Technology.

The aim of this work was to find the best possible detector for in situ measurements of low energy gamma radiation in close geometries. In situ gamma spectrometry is an important and reliable method after a radionuclide deposition, e.g. after a nuclear power plant accident. Details were gathered of currently available detectors that have high efficiency for low energy gamma radiation. Following this, an investigation was made to optimize detector parameters, i.e. to find parameters that provide the best measurement results from a sensitivity point-of-view. This was possible using Monte Carlo calculations where e.g. crystal and window material, crystal size and distance between crystal and window can be varied to find the best combination. A larger detector crystal provides a higher efficiency and higher peak-to-Compton ratio. In a small detector, it is more likely that the gamma ray will Compton scatter out of the detector instead of depositing all of its energy. A thinner window made of a material of low density and low atomic mass is of more importance for low energies, where beryllium is commonly used. Semi-planar detectors offer high resolution and excellent efficiency up to a few hundred keV.

Log: 424. **PHYSIOLOGICAL EXPERIMENT TOWARD THE REDUCTION OF RADIOCAESIUM CONTAMINATION IN A RICE PLANT.** Kobayashi, N.I. (1); Nobori, T (1); Tanoi, K (1); Nemoto, K (1); Nakanishi, T.M. (1). (1) the University of Tokyo.

After the accident of the Fukushima nuclear power plant in 2011, a lot of attention has been paid particularly to the radiocaesium content in a rice plant in Japan. To ensure the low radioactivity in the rice grain, it is important to define the factors affecting the radiocaesium behavior in a rice plant. The survey data collected from various paddy fields in Fukushima clearly demonstrated that soil potassium (K) content negatively correlated to the radiocaesium content in a rice plant, resulting in the encouragement of K fertilization. However, the scientific basis for the effect of K supply on the radiocaesium uptake and transport is still insufficient. Therefore, we carried out Cs-137 tracer experiment to investigate the physiological effect of K supply on the radiocaesium uptake and movement. When the rice plant was grown in the solution containing 3 mM K for 8 weeks, Cs-137 content in the shoot was reduced to less than half of that grown under K deficient condition. It was also found that the amount of Cs-137 transferred from old leaves to new leaves and ears was significantly decreased by supplying K in the solution. In the case of the grain, Cs-137 content was reduced to one-sixth when K was supplied. These results were in good accordance with those in the field experiment, i.e. comparing the radiocaesium content in rice plants grown with or without K fertilization. Therefore, K concentration in soil was suggested to be the major factor affecting the radiocaesium content in rice grain.

Log: 425. **DEGRADATION OF LI-ION BATTERIES: AGING STUDIES USING THE PGAA TECHNIQUE.** Kudejova, P.(1); Seidlmayer, S.(1); Buchberger, I.(2); Bernhard, R.(2). (1) MLZ, Technische Universitaet Muenchen. (2) Technical Electrochemistry, Technische Universitaet Muenchen.

The broad use of the Li-ion batteries in electronic devices of our everyday life increases the requirements on the quality of the batteries. Long-term recharging leads inevitably to degradation of batteries by fade in their capacity. It is suspected that the process is connected with the dissolution of the transition metal ions of the cathode and their deposits on the anode material. The PGAA method was used to qualitatively determine the deposition of Mn, Ni, Co (in LiNi<sub>1/3</sub>Mn<sub>1/3</sub>Co<sub>1/3</sub>O<sub>2</sub> battery type) and Fe (in LiFePO<sub>4</sub> battery type) on the anode material. The anodes were made of ca. 10 mg graphite – an ideal matrix for PGAA measurement and analysis. The typical backing of the anode consists of ca. 8 mg thin pure copper foil. The copper signal was used for comparison as an internal standard. The evaluation of Mn concentration was cross-checked using the decay spectra of Mn with respect to the decay peaks of the copper backing as well.

Log: 426. **MONITORING INSPECTION FOR RADIOCESIUM IN AGRICULTURAL, LIVESTOCK, FORESTRY AND FISHERY PRODUCTS IN FUKUSHIMA PREFECTURE.** NAOTO NIHEI(1); KEITARO TANOI(1); TOMOKO M. NAKANISHI(1). (1) THE UNIVERSITY OF TOKYO.

Radioactive nuclides, including radiocesium, emitted from the nuclear accident at Fukushima in 2011 contaminated agricultural land in Fukushima Prefecture. To keep the agricultural products in safe, the Ministry of Health, Labor and Welfare established a tentative regulation level of 500 Bq/kg for radiocesium in cereals, vegetables, meat, and fishery products. In April 1, 2012, a new value of 100 Bq/kg was introduced to regulate radiocesium concentration in general food. In the present study, we summarized the inspection results of radiocesium concentration in the products of Fukushima Prefecture during 3 years after the accident. The ratios of radiocesium concentration that exceeded the limit of 100 Bq/kg from March 2011 to June 2011 were 18% in agricultural products (excluding rice), 3% in livestock productions, 49% in forest productions, and 52% in fishery products. After June 2011, radiocesium concentration reduced drastically. Radiocesium concentration in agricultural and livestock products hardly exceeded 100 Bq/kg. Radiocesium concentrations of forest and fishery products have been decreasing every year, but there were a few samples which exceeded 100 Bq/kg. Rice is one of the most important agricultural products in Japan. Therefore, in 2012, Fukushima Prefecture decided to measure the radiocesium concentration of all the rice produced in Fukushima, using custom-made belt conveyor testers. The contaminated rice detected by the inspection was reduced year by year. The rice with radiocesium concentration over 100 Bq/kg were detected in only 71 bags out of 10,338,000 bags in 2012 and 28 bags out of 11,001,000 bags in 2013.

Log: 427. **CERTIFIED REFERENCE MATERIALS OF AGRICULTURAL PRODUCTS AND FOODS BEARING RADIOACTIVITY DERIVED FROM FDNPP.** Minai, Y.(1,2); Miura, T.(3); Yonezawa, C.(4); Iwamoto, H.(5); Shibukawa, M.(6); Takagai, Y. (7); Furukawa M.(8); Arakawa, F.(9); Okada Y.(10); Kakita, K.(11); Kojima, I.(11); Hirai, S.(10). (1) Musashi University, (2) Nedzu Chemical Laboratory, (3) NMIJ/AIST, (4) CPDNP/JIIA, (5) Environmental Technology Service Co. Ltd., (6) Saitama University, (7) Fukushima University, (8) Perkin-Elmer Japan Co. Ltd., (9) Nippon Meat Packers Co. Ltd., (10) Tokyo City University, (11) JSAC.

Preparation of reference materials for determining radioactivity in agricultural products and foods has been an emergent task in Japan after FDNPP. Our team supported by Japan Science and Technology Agency (JST) has conducted the program developing the reference materials with the certification by Japan Society for Analytical Chemistry (JSAC) since early summer in 2012. The first CRM was prepared from brown rice collected at a village in Eastern Japan. No radioactivity is promoted in any preparation scheme involved. Complete mixing of the rice granules achieved satisfactory homogeneity for determining Cs-134 and Cs-137. Over 10 domestic institutions joined the round-robin test for evaluating the radioactivity in the CRM candidate. After certification for radioactivity of Cs-134, Cs-137, and natural K-40, the CRM of brown rice has been distributed before the harvest season in 2012. Applying similar procedures, the following CRMs having Cs-134 and Cs-137 derived from FDNPP were developed: soybean powder, 'shiitake' mushroom powder, and beef flake. Concentration of total radiocesium were around several hundreds Bq/kg. The latest CRMs developed in this program are prepared from fish samples taken from the Fukushima coast. Each marine fish sample (Okamejei and kenojei) was separated into two portions: bone and meat parts. Strontium-90 could be enriched in the bone part whereas radiocesium would be distributed in both parts. The fish bone CRM for Sr-90, Cs-134, and Cs-137 and the fish meat CRM for Cs-134 and Cs-137 would be distributed in the spring,

Log: 428. **INTERNATIONAL INTER-COMPARISON EXERCISE ON GAMMA-RAY SPECTROPHOTOMETRY FOR CERTIFIED REFERENCE MATERIAL OF BROWN RICE BEARING FDNPP-DERIVED RADIOCESIUM.** Yonezawa, C.(1); Minai, Y.(2,3); Hirai, S.(4); Miura, T.(5); Kakita, K.(6); Kojima, I.(6); Iwamoto, H.(7); Okada, A.(8); Okada, Y.(4); Arakawa, F.(9); Kitamura, K.(10); Sanada T.(11); Yamada, T.(12); Shibukawa, M.(13); Chiba, K. (5); Uematsu, Y.(14). (1) JIIA, (2) Musashi Univ., (3) Nedzu Chem. Lab., (4) Tokyo City Univ., (5) NMIJ/AIST, (6) JSAC, (7) Env. Tech. Serv., (8) Toshiba Env. Sol., (9) Nippon Meat Packers, (10) JCAC, (11) Hokkaido Univ. Sci., (12) JARA, (13) Saitama Univ., (14) JAB.

Preparation of CRM of brown rice granule contaminated with radiocesium derived from FDNPP was completed in the summer of 2012. Distributing the CRM before the harvest season was the emergent task for the development group. It was decided to carry out the round-robin test by the domestic institutions promising quick response for the request on radiometric analysis of the CRM candidate. This resulted in necessity of inter-comparison study involving the foreign institutions recognized in the international community for ensuring reliability of the CRMs developed. The brown rice CRM was chosen for this purpose because that was the most popular product we developed after FDNPP. Six institutions (HMGU/ISS, Germany; IRMM, EU; IAEA-TEL, international; NIST, U.S.A.; NPL, UK; PTB, Germany) participates the inter-comparison exercise; the details on the exercise will be published elsewhere by the international members from the participating institutions and the CRM-developing team. This paper presents the preliminary results summarized by the Japanese team having the development task for the CRMs after FDNPP. The mean value of concentration for each nuclide (Cs-134, Cs-137, or natural K-40) agreed with the certified value based on the data from the domestic laboratories. The relative difference between the mean value and the certified value based on the previous data was less than 2.3%, which corresponds to homogeneity of radioactivity in the CRM.

Log: 429. **PREPARATION OF RADIOACTIVE TELLURIUM AND ESTIMATION OF ITS MIGRATION BEHAVIOR IN SOIL - PLANT SYSTEM.** Fukutani, S.(1); Takahashi, T.(1); Fujiwara, K.(1); Ikegami, M.(1); Kinouchi, T.(1); Fujii, T.(1). (1) Kyoto Univ.

The Fukushima nuclear power plant issue caused widespread radioactive contamination of various nuclides. Radioactive tellurium (Te) was one of the various nuclides released from the nuclear power plant, and Te-132 and Te-129m were detected in atmospheric, soil, and freshwater product samples. Te-127m is one of radioisotopes of tellurium, and its release amount was estimated to be  $1.1 \times 10^{15}$  Bq. Te-127m has a relatively long half-life of 109 days, and was considered to be an important nuclide for internal exposure at the initial stage of accident. In this study, we prepared radioactive tellurium from stable tellurium by neutron irradiation and activation at research reactor, and estimated migration behavior of radioactive tellurium added into soil - plant system.

Log: 430. **THE IMPACT OF THE EXTENSIVE USE OF PHOSPHATE FERTILIZERS ON RADIOACTIVITY LEVELS IN FARM SOIL IN TANZANIA.** Mohammed, N. K. (1); Chanai, E. E. (1); Alkhorayef, M. (2)(3). (1) University of Dar es Salaam. (2) King Saud University. (3) University of Surrey.

The use of different types of fertilizers including phosphates in agricultural activities in Tanzania is increasingly becoming important in recently years. The practice is motivated by the Government through fertilizer subsidies made all over the country for the purpose of bringing green revolution. The most commonly used phosphate fertilizers in Tanzania are single superphosphate (SSP), di-ammonium phosphate (DAP) and triple superphosphate (TSP), which are imported from outside the country, as well as a local phosphate fertilizers from Minjingu phosphate deposits in Arusha which is a raw phosphate rock crushed into powder and granules. However, despite the enormous benefits derived from the use of fertilizer, literature has shown that phosphate fertilizers are normally associated with elevated concentrations of U-238, Th-232 and their progenies such as Ra-226 and Ra-228. On top of that the Minjingu phosphate rock is reported to contain higher activity concentration of U-238 and its progenies ( $\sim 4000$  Bq/kg) than many phosphate rocks reported elsewhere. Therefore, there is a need to assess the activity concentration of radionuclides associated with phosphate fertilizers in farm soil. In this study samples of soil from farms in Iringa rural were analysed for radioactivity using gamma ray spectrometry of the Tanzania Atomic Energy Commission in Arusha Tanzania. The study was conducted in Iringa rural because it is one of the districts with a combination of farms that make extensive use of phosphate fertilizers and farms that grow similar vegetables organically. The aim of this work was to dertermine the radioactivity levels in soil associated with the use of phosphate fertilizer in farming. The informations obtained can be used by the authority to evaluate the radiological risks associated with the use of phosphate fertilizers in agricultural activities.

Log: 431. **DETERMINATION OF RADON GAS IN NATURAL GAS PIPELINES.** Chanyotha, S.; Kranrod, C.; Phangvanich, P.; Sriploy, P. Chulalongkorn University. The aim of this study was to develop the methodology for collection and analysis of radon gas from a natural gas pipeline. We used activated charcoal as an adsorber material. Two methods were designed for collecting radon gas samples. For onshore case, a continuous gas sampling method from the pipeline was preferred. However, the discharge point where the charcoal adsorber is deployed needs to be in a safe area. For offshore cases, a batch sampling method was designed. The gas sample was collected in an industry-approved sample container (bomb) capable of direct sampling from high-pressure (800 to 2000 psi) distribution lines, and then transferred to the offshore laboratory facility to be discharged into the charcoal adsorber. The volume of gas sample required for analysis should be more than 18-20 liters with a sampling flow rate of 2-4 liter/sec. A gas regulator was used to control the flow rate of the gas in order to estimate the volume of the gas that passed through the charcoal canister. Gamma spectroscopy technique was utilized to determine the concentration of radon by analysis of radon daughters on the charcoal at the NORM laboratory, Chulalongkorn University. These gas collection techniques have been calibrated using the radon standard chamber.

Log: 432. **RADIOCESIUM BEHAVIOR IN A PEACH TREE AFTER THE FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT (FDNPP) ACCIDENT.** Tanoi, K. (1); Takata, D. (1); Kobayashi N.I. (1); Hirose A. (1); Nakanishi T.M.(1). (1) The University of Tokyo. Radiocesium contamination on agricultural fields has been a great concern after the FDNPP accident. The soil-to-plant transfer factor of radiocesium in fruit trees was low enough to be optimistic about contamination in fruits. However, in the monitoring inspection process, some fruit species including peach contained more than 100 Bq/kg radiocesium (expressed as the sum of Cs-134 and Cs-137). To study the contamination mechanism, we investigated radiocesium distribution in peach trees grown in the orchards. First, we visualized the radiocesium in the bark, since the trunk was the only part directly contaminated by the fallout because the peach tree had no leaves or flowers when the accident occurred in March. The radiocesium was detected in uneven parts of the bark as well as around the lenticel, connected to the cambium layer, leading to the vascular bundle. It was found that the radiocesium concentration in fruits grown at the covered soil was the same as those grown in the open place. Combined with data showing that radiocesium concentration in the vascular bundle was higher than in the inner bark it was suggested that radiocesium attached on the bark was straightforwardly moves toward the inner vascular tissues of the peach tree. This study shows that the radiocesium uptake from the root was very small compared to the transfer through the bark-lenticel route.

Log: 433. **Chemical durability and structural analysis of Na<sub>2</sub>O-PbO-B<sub>2</sub>O<sub>3</sub> and Na<sub>2</sub>O-PbO-P<sub>2</sub>O<sub>5</sub>-B<sub>2</sub>O<sub>3</sub> glasses and testing leaching rate for simulated radioactive wastes.** Sema Akyil Erenturk(1), Murat Bengisu (2), Cem Erdogan (3) (1) Istanbul Technical University, Energy Institute, 34469 Maslak, Istanbul, Turkey (2) Izmir University of Economics, Department of Industrial Design, Sakarya Cad., No. 156, 35330 Balçova, Izmir, Turkey (3) Ege University Institute of Nuclear Sciences, 35100 Bornova, Izmir, Turkey.

The present study is concerned with the development of new borate glasses for the immobilization of nuclear waste and optimization of these glasses to attain homogeneous glass structure, high chemical durability, high strength, and minimized leaching of radioactive elements to protect environment. We aim to develop alternative glass compositions resulting in waste products with high chemical durability, homogeneous structure and lower processing temperatures than borosilicate glasses used for the same purpose. Furthermore, the addition of PbO to the composition is investigated due to its capacity to block gamma radiation from the immobilized mass to the environment. PbO is also of interest due to its capacity to lower the melting temperature and increase the chemical durability of the glass. Technical procedures have been developed to eventually apply such compositions for the immobilization of cesium- and strontium-containing HLW. However at this early stage of the long-term research project we experiment with simulated waste for practical and safety reasons. The results suggest that such compositions are potential candidates for feasible hosts for nuclear waste immobilization to avoid leaching of the radionuclides to environment. Various sodium borate glass compositions were initially tested with respect to glass formation ability, chemical durability and leaching rate of radionuclides in vitrified nuclear waste.

Log: 434. **TOENAIL ELEMENTAL ANALYSIS OF KOREAN ADULTS BY INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS.** Lee O(1), Moon J H(2), Kim S. H.(2), Chung Y. S.(2), Sun G.(2) (1) Yongin Univ. Dept of Food Science and Nutrition (2) KAERI NAA lab.

Mineral elements are constituents of body skeleton and fluids, and act as functional components for many biochemical processes in human body. Their concentrations in body tissue provide useful information about health status. Elements such as Se and Zn play important roles for the maintenance of anti-oxidative capacity and immunity in human. The elemental contents in toenail represent for long-term exposure of some minerals. The toenail is non-invasive human sample. Many countries reported diverse toenail elemental contents in their populations. Instrumental neutron activation analysis was engaged to measure the toenail mineral contents in Korean young adults using research reactor. Toenail samples collected from young men and women were analyzed and the concentration level of the elements were evaluated. NIST SRMs were analyzed simultaneously for an analytical quality control. The average toenail Na and K contents were 643 and 817 ppm, Ca was 1368ppm and Fe was 644ppm. Besides, toenail Se, Zn, and Mn have a concentration level of 0.77, 99.6 and 23.2 ppm, respectively



Log: 435. **Elemental characterization of lignite from Afsin-Elbistan in Turkey by k0-NAA.** Marie Kubešová (1), Esra Orucoglu (2), Sevilay Hacıyakupoglu (3), Sema Erenturk (3), Ivana Krausová (1) (1) Nuclear Physics Institute ASCR, 250 68 Řež, Czech Republic (2) Faculty of Mines, Istanbul Technical University, 34469 Maslak, Istanbul, Turkey (3) Energy Institute, Istanbul Technical University, 34469 Maslak, Istanbul, Turkey.

As a result of industrialization and population increase, the energy demand rises in the world and also in Turkey. Although coal has some deficiencies, it still presents a major source for energy need. Due to mining activities or coal combustion, enrichment of some elements like uranium, arsenic, selenium in coal and ash compared with the host rocks can cause environmental pollution. Therefore accurate determination of concentration levels of hazardous elements in coal is important. Elemental analysis of coal is also important for the economic perspective as it helps to understand the inorganic geochemistry of lignites and/or evaluate a possibility recovery of some valuable elements from its ash. The Afsin-Elbistan lignite deposit is located in the southeast of Turkey in Kahramanmaraş Province within Afsin and Elbistan districts. Around 95 thousand people live there in the close proximity of the energetic complex which consists of two lignite-fired power plants Afsin-Elbistan A and Afsin-Elbistan B with total power of 2795 MW. Although the quality of the deposit is low with an average calorific value of 1070 kcal/kg the deposit is one of the most important source of electricity generation in the country. 12 samples of lignite taken from several places and depths of the Kıslaköy open cast mine in Afsin-Elbistan were characterized by k0-NAA with emphasis on the potentially hazardous elements, such as Al, As, Br, Cd, Cl, Cr, Cu, Fe, I, Mn, Ni, S, U, V, Zn, and others (62 elements in total). The analysis showed low quality and elemental inhomogeneity of the lignite material, which is used in the nearby thermal power plant. The results were compared also to the older data from the same locality.

Log: 436. **THE DEFENSE THREAT REDUCTION AGENCY'S TECHNICAL NUCLEAR FORENSICS MATERIAL COLLECTIONS, ANALYSIS, AND DEBRIS DIAGNOSTICS R&D PROGRAM.** Feener, J.S.; Morin, C.L.; Zickafoose, M.S. DTRA.

The Defense Threat Reduction Agency (DTRA) Technical Nuclear Forensics Material Collections, Analysis, and Debris Diagnostics Research and Development (R&D) Program's overarching goal is to develop targeted technologies, methodologies, and tools to enable the collection, analysis and interpretation of the composition of detonation materials relevant to the nuclear forensics assessment and policy decision maker community within a designated timeframe after the initial event. This goal is attained through the Materials Identification and Debris Analysis Solutions (MIDAS) Series strategy with four subgoals: (1) reduce fixed laboratory analysis timeline in the current construct by 50% or more; (2) produce a field analysis and collections kit for priority elements as defined by assessment needs and have a means to interpret the analysis results; (3) develop technologies, materials, exercises, tests, or models that enable enhanced verification and validation (V&V) of new analysis and methodologies; and (4) develop revolutionary technologies to radically transform the way nuclear forensics is performed. To accomplish these goals, DTRA funds R&D projects focused on: (1) advancing the traditional construct for traditional answers; (2) development of a new paradigm / new conduct of operations; (3) enhanced V&V including testing and improved materials and models; and (4) innovative, game-changing technologies. This paper will outline the MIDAS Strategy and current R&D interests with the intent of reaching the larger radioanalytical chemistry and radiochemistry community represented at the International Conference on Methods and Applications of Radioanalytical Chemistry.

Log: 437. **FISSION PRODUCT COLLECTION AND PURIFICATION FROM THE SPONTANEOUS FISSION OF CF-252.** Pfeiffer, J. K. (1); McGrath, C. A. (2); Carney, K. P. (1). (1) Idaho National Laboratory. (2) Idaho State University.

With the sparse availability of fast fission sources in the United States, the development of methods to produce isotope mixtures emulating the fission of uranium and plutonium using a fast spectrum presents an interesting research challenge. One method that INL is pursuing is the collection of isotopes from the spontaneous fission decay of Cf-252. The advantage of this approach is that isotopes can be produced on demand without the need for fast critical assemblies or reactors. A planchet with electroplated Cf-252 is contained in a source holder. A thin nickel foil separates the californium source from the collection target; fission fragments transmit through the foil while heavier isotopes (californium and decay daughter curium) are blocked. Fission fragments are implanted onto a metallic target. After an appropriate amount of time, the target is removed from the source holder and fission products are leached into an acidic solution. An isocratic HPLC method for separating lanthanides has been characterized using a stable surrogate solution containing yttrium, and lanthanum through terbium. This separation was performed with good repeatability (generally < 3% RSD); most elements were isolated in fractions with purity greater than 93% and yields exceeding 90%. The separation of elements can be tailored by varying eluent pH, resulting in predictable changes in element elution order and retention time. This was used to optimize purity and yield for elements that co-eluted at a certain solution pH. This method will be applied to the separation and analysis of fission products collected from the spontaneous fission decay of Cf-252.

Log: 438. **A SIMPLE AQUEOUS APPROACH TO THE SYNTHESIS OF NEPTUNIUM IODIDE.** Pfeiffer, J. K. (1); Johnson, A. T. (1); Martin, L. R. (1); Carney, K. P. (1); Finck, M.R. (1). (1) Idaho National Laboratory.

Researchers at INL have been working on the synthesis of volatile neptunium compounds in order to improve the sensitivity of mass spectrometric measurements. One approach being considered is the development of neptunium halide complexes. Naturally occurring chlorine and bromine are multi-isotopic, resulting in complications in the mass spectrum due to the numerous isotopes existing in the counter ions. Although mono-isotopic fluorine is spectrometrically simpler, it is highly corrosive. Production of neptunium iodide is advantageous because it is more volatile than chloride and bromide counterparts and is mono-isotopic. Two aqueous methods for the production of Np(IV) iodide have been developed. The first utilizes controlled potential electrolysis of neptunium in nitrate media for reduction to Np(IV). Np(IV) is extracted into Thenoyltrifluoroacetone (TTA) in toluene, then back extracted into hydroiodic acid. The second method uses controlled potential electrolysis to reduce Neptunium in iodide media, generating Np(IV) iodide in situ. UV-visible-nIR spectroscopy analysis was used for initial confirmation of successful synthesis of neptunium tetra-iodide species via both routes. Solid neptunium tetra-iodide will be induced by crystallization, enabling further investigations into the crystal structure, thermodynamic properties and vapor pressure of this material. Techniques for synthesis of the similar Neptunium tri-iodide compound and characterization will also be discussed.

Log: 439. **EXPLORING RAPID RADIOCHEMICAL SEPARATIONS AT THE UNIVERSITY OF TENNESSEE RADIOCHEMISTRY CENTER OF EXCELLENCE.** Hall, H.L. and Auxier II, J.D., University of Tennessee.

The University of Tennessee formed its Radiochemistry Center of Excellence (RCOE) in 2013 with support from the U.S. National Nuclear Security Administration. One of the major thrusts of the RCOE is to develop deeper understanding of rapid methods for radiochemical separations that are relevant to both general radiochemical analyses as well as post-detonation nuclear forensics. Early work has included the development and demonstration of rapid separations of lanthanide elements in the gas phase, development of a gas-phase separation front-end for ICP-TOF-MS analysis, and the development of realistic analytical surrogates for post-detonation debris to support methods development. In addition, the RCOE is training new students in the important field of radiochemistry -- helping to fill a significant gap in developing new expertise and practitioners. This paper reviews the early work and accomplishments of the RCOE and highlights the general research directions in radiochemical separations that are being pursued.

Log: 440. **DEVELOPMENT OF RADON-IN-WATER AND RADON-IN-AIR REFERENCE MATERIALS.** Lee, K.Y. (1); Kim, M.J. (2); Cho, S.Y. (1). (1) Korea Institute of Geoscience and Mineral Resources. (2) Neosiskorea Co., Ltd.

Because radon (Rn-222) has a short half-life of 3.8 days, the commercial reference material has not been manufactured considering the long stability and its matrix matching. In this study, we developed the radium (Ra-226) free radon reference materials using self-developed radon-generating-system (RGS). RGS is composed of radon source container (solid or liquid material containing radium is stored), radon detector, radium removal unit, drying column and a few dispensing vessels. Radon which generated from radium in the radon source container is evenly distributed through the RGS closed-loop by an air pump inside the radon detector. After radon is fully distributed, the dispensing vessels in the closed-loop are separated. The radon concentration of each dispensing vessel can be exactly evaluated from the known volumes of each component and whole system. In addition, the vessel containing air or water can be used as the radon reference material for gas or liquid sample, respectively. In the present condition, the measurement accuracy both radon concentration and closed-loop volume agreed well within 10% error range.

Log: 441. **Advances in Np-237 Mossbauer Spectroscopy.** Lucas E. Sweet, David G. Abrecht, Benjamin S. McDonald, Dallas D. Reilly, Erin S. Fuller, Ravi K. Kukkadapu, Jon M. Schwantes.

Actinide Mossbauer spectroscopy was developed nearly 50 years ago, yet today there are few labs in the world actively performing this chemical characterization technique. Mossbauer spectroscopy is known to be a powerful chemical speciation technique requiring little or no sample preparation. However, some of the issues limiting the utility of actinide Mossbauer spectroscopy are the sensitivity, measurement times and cost of operation. Recent advances in instrumentation used for iron Mossbauer spectroscopy led us to examine how actinide Mossbauer spectroscopy might be enhanced to be more readily usable. This work is focused on an improved Np-237 Mossbauer spectrometer design that increases the utility of this chemical characterization technique for basic and applied science.

Log: 442. **INTEGRATING MICROFLUIDICS FOR THE MINIATURIZATION OF NUCLEAR MATERIAL ANALYSIS TECHNIQUES.** Benjamin T. Manard (1), Xu, Ning (1), Gao, Jun (1), McCulloch, Quinn (1), Chamberlin, Rebecca M. (1), Montoya, Dennis Patrick (1), Castro, Alonso (1). (1) Los Alamos National Laboratory.

Miniaturization has taken analytical chemistry by storm over the past two decades ranging from lab-on-a-chip devices (micro total analytical systems,  $\mu$ TAS) to palm sized sensors and spectrometers. Unfortunately, little work has been dedicated for actinide chemistry, particularly separations. Presented here is an innovative technique that employs a microchip for the processing of actinide separations. Commonly for trace metal analysis of nuclear materials, inductively coupled plasma optical emission spectroscopy (ICP-OES) is employed for detection due to its sensitivity, robustness, and rapid sample analysis time. However, emission line rich plutonium (Pu) and uranium (U) in sample matrix can hinder the determination of trace metals due to spectral overlap. To correctly quantify the trace elements, Pu and/or U must be removed from the sample prior to ICP-OES analysis. The primary goal of this study is to develop a separation technique by employing a microchip to remove Pu/U for analysis of trace elements such that sample size and radiological waste is reduced. LA-UR-14-29310.

Log: 443. **Partitioning processes for minor actinides: influence of gamma radiation on extraction systems based on diglycolamides.** Galán, H.(1); Núñez, A.(1); González, A.(1); Cobos, J.(1); ; Durana, A.(2); de Mendoza, J.(2); Egberink, R.J.M.(3); Verboom, W.(3); Munzel, D.(4), U. Müllich(4), A. Geist (4). (1) Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas, Madrid, Spain. (2) Institut Català d'Investigació Química (ICIQ), Tarragona, Spain. (4) University of Twente, Enschede, The Netherlands, (4) Karlsruhe Institute of Technology (KIT), Germany.

For advance nuclear fuel cycles, different approaches based on extraction processes are being developed for minor actinides recycling contained in high level radioactive waste. To predict the behaviour along the extraction process is one of the most important challenges in defining the process development and safety. Solvents used for the partitioning will be in contact with highly radioactive aqueous solutions containing elevated nitric acid concentrations, therefore, it must be identify any unexpected behaviour as well as maximize the regeneration of the spent solvent. Malonamides and diglycolamides are considered as the extractants with higher possibilities to be used at large-scale in Europe. This work describe the physico-chemical effects of gamma radiation on solvent systems based on TODGA (N,N,N',N'-tetraoctyl diglycolamide) such as the DIAMEX, i-SANEX and GANEX processes. The behaviour of the extraction systems under radiation, by using a homogeneous gamma flux of external  $^{60}\text{Co}$  sources, has been explored depending on the structural stability of ligand, the nature of the diluents or the presence of different ions, etc.. A systematic characterization of degraded samples (composition and extraction properties) and the study of their interferences with other phases are being carried out.

Log: 444. **SIMULATING TRANSIENT SITUATION IN THE SUBCRITICAL REACTOR USING A SEALED-TUBE NEUTRON GENERATOR.** Li, G.(1); Shi,Y.Q.(2); Luo, H.D.(2); Liu, Z.(1); Liu, L.M.(1). (1) Northeast Normal University. (2) China Institute of Atomic Energy (CIAE).

The characteristic of transient situation is one of the frontier researches in the field of accelerator driven subcritical reactor (ADS). The operation of high power proton accelerator (HPPA) is complex and unrealistic to adjust the parameters of HPPA frequently. A sealed-tube neutron generator designed by northeast normal university (NENU) on the physics test platform of ADS in China institute of atomic energy (CIAE) is used to develop simulation test system of reactivity. The neutron source in a simulation of transient situation is required to generate changed neutron flux which is fast, random, and continuous. Simple pulsed neutron can not meet above requirements. It is possible to simulate transient situation by modulating a pulsed neutron of high frequency due to a large time constant of reactors. By improving a driver of neutron tube ion source and modulating a trigger signal of pulsed neutron, the simulating transient situation of HPPA is done which includes the beam forward transient situation, beam trip, beam step increase/decrease, and beam shutoff. In practices, this method has been used to do the preliminary test of a recovery characteristic under transient situation in a subcritical reactor. The results are well coincident with the theoretically calculating results. This method is more accurate, fast, and reliable. It is also more convenient to simulate the transient situation of HPPA, more important to do research of impact of transient situation on ADS, and more useful to improve the ADS.

Log: 445. **SIMULATED BETA-GAMMA DATA FOR VERIFICATION AND VALIDATION, TRAINING AND DATA INJECTS INTO THE NATIONAL DATA CENTERS EXERCISES.** McIntyre, J.(1); Schrom, B.(1); Suckow, T.(1); Prinke, A.(1); Ringbom, A.(2). (1) Pacific Northwest National Laboratory. (2) FOI Defence Research Establishment.

Several hundred simulated radioxenon beta-gamma data files were developed to assist in evaluating the performance and results from radioxenon concentration calculation analysis. After an initial program was developed, additional uses began to be identified for the program output: training sets of two-dimensional spectra for data analysts at the International Data Center (IDC) and other National Data Centers (NDC), spectra for exercises such as the Integrated Field Exercise 2014 (IFE14) at the Dead Sea in Jordan, and to test new analysis methods and algorithms. PNNL developed a Beta-Gamma Simulator (BGSim ) that incorporated GEANT modeled data sets from radioxenon decay chains, as well as functionality to use nuclear detector acquired data sets to create new beta-gamma spectra with varying amounts of background, Xe-133, Xe-131m, Xe-133m, Xe-135 and Rn-222 and its decay products. The program has been implemented on a Web-based applications platform and allows the user to create very specific data sets that incorporate most of the operational parameters for the current beta-gamma systems deployed in the International Monitoring System (IMS) and the On-site Inspection (OSI) equipment.

Log: 446. **STUDY OF EXCHANGE NETWORKS BETWEEN TWO AMAZON ARCHAEOLOGICAL SITES BY INAA.** Hazenfratz, R. (1); Munita, C. S. (1); Neves, E. G. (2) (1) Nuclear and Energy Research Institute (Sao Paulo/Brazil). (2) Museum of Archaeology and Ethnology of the University of Sao Paulo.

This work focuses on an archaeometric study involving archaeological ceramic material from two large archaeological sites in Central Amazon, namely Lago Grande and Osvaldo, on the confluence region of Negro and Solimões rivers. It was tested a hypothesis about the existence of an exchange network between the former inhabitants of those sites, focusing on material and/or technological exchange. That hypothesis has implications for archaeological theories of human occupation of the pre-colonial Central Amazon, which try to relativise the role of ecological difficulties of the tropical forest as a limiting factor for the emergence of social complexity in the region. The physical-chemical characterization of potsherds and clay samples near the sites was carried out by instrumental neutron activation analysis (INAA) to determine the elemental chemical composition. Previous studies showed that Osvaldo and Lago Grande were occupied by people who produced pottery classified in the Manacapuru and Paredão phases, subclasses of the Incised Rim Tradition, around the 5-10th and 7-12th centuries BC, respectively. INAA results were analyzed by multivariate statistical methods, whereby two chemical groups of pottery were defined for each archaeological site. By integration of the results with archaeological data, the superposition between pairs of chemical groups was interpreted as a correlate of an ancient exchange network, although it was not possible to define if it existed exclusively between Lago Grande and Osvaldo. On the contrary, it was suggested that Lago Grande participated in a more extensive exchange network by comparison of two chemical groups.



Log: 447. **FUSION OF AERIAL GAMMA RAY SURVEY AND REMOTE SENSING DATA FOR A DEEPER UNDERSTANDING OF RADIONUCLIDE FATE AFTER RADIOLOGICAL INCIDENTS: EXAMPLES FROM THE FUKUSHIMA DAI-ICHI RESPONSE.** McLean, L.K.(1); Czaja, W.C. (2); DiBenedetto, J.A. (3). (1) Remote Sensing Laboratory. (2) University of Maryland. (3) Special Technologies Laboratory.

Aerial gamma ray surveys can be used to characterize the scope of large radiological releases. The intensity of the radiation field can be mapped with spatial detail that varies according to the constraints placed on the flight parameters by factors such as the equipment availability and terrain complexity. Although limited quantitative information can be obtained via spectroscopy, the quality of the data doesn't support rigorous calculations of the isotope-specific deposition. To address these challenges as experienced in the DOE response to the Fukushima Dai-ichi incident, non-linear dimension reduction methods have been developed that enable the clustering of the data into coherent spectral groups. These methods enable qualitative assessments of the spectral content and are more robust against the difficulties associated with aerial survey data. The results of the application of these methods to the data from Fukushima show strong spatial coherence and reveal subtle and isolated spectral signatures. Furthermore, the fate of radionuclides following an atmospheric release is dependent upon to the surfaces onto which they are deposited. By fusing the gamma ray data with optical and electro-optical data, land use and other meta data can be associated with specific gamma ray signatures. This can provide insights into the preferential retention, dissipation or movement of radionuclides in specific environments. The incorporation of remote sensing data into the aerial gamma ray mapping mission has significant potential impact on the consequence management mission as well as post-detonation nuclear forensics and non-proliferation.

Log: 448. **SPECTROMICROSCOPIC TECHNIQUES FOR NUCLEAR FORENSICS.** Ward, J.D.(1); Duffin, A.M.(1); Eiden, G.C.(1). (1) Pacific Northwest National Laboratory.

Spectromicroscopic techniques that measure the structure and chemical composition of materials on a submicron spatial scale are a potentially powerful complement to currently available nuclear forensics techniques. For example, scanning transmission X-ray microscopy (STXM) beamlines available at advanced synchrotron facilities focus soft X-rays to a small (~10 nm) spot size. This, in conjunction with the fact that soft X-ray absorption spectra are highly sensitive to the physical and chemical structure of the analyte, enables one to collect chemical information at a very fine spatial scale. This talk will demonstrate how STXM can be used to perform chemical imaging of heterogeneous samples at high spatial resolution. In addition, we will demonstrate the use of STXM to study the chemical ageing of reactive uranium materials. This physical and chemical imaging can reveal information about the sample not available to techniques which measure the isotopic composition alone.

Log: 449. **SPATIAL ISOTOPIC URANIUM ANALYSIS WITH FS-LA-MC-ICPMS.** Andrew M. Duffin; Jesse D. Ward; John W. Robinson; Mackenzie C. Endrez; Gregory C. Eiden. Pacific Northwest National Laboratory.

Femtosecond laser ablation coupled with multicollector inductively coupled plasma mass spectrometry (fs-LA-MC-ICPMS) has proven a powerful tool for uranium isotopic analysis. The main advantages of this technique include limited or no sample preparation, excellent precision and sensitivity, and rapid sample analysis. In addition, the focused ablation laser allows high precision (ca. 10 micron) spatial analysis, providing a means to isotopically map a heterogeneous sample. In this report, we will present the performance characteristics of fs-LA-MC-ICPMS for spatial analysis and show isotopic maps of samples prepared to test the spatial resolution and sensitivity of fs-LA-MC-ICPMS.

Log: 450. **ANALYSIS OF NON-TRADITIONAL RADIOXENON ISOTOPES.** Klingberg, F.J.(1); Biegalski, S.R. (1); Prinke, A. (2); Haas, D. (2); Lowrey, J. (2).

As part of the verification component of the Comprehensive Nuclear-Test-Ban Treaty (CTBT), the four xenon isotopes Xe-131m, Xe-133m, Xe-133, and Xe-135 originating from nuclear fission are traditionally analyzed. In this work, the non-traditional radioxenon isotopes Xe-127, Xe-127, Xe-129m, and Xe-137 were investigated. The isotopes were produced in isotopically pure samples via neutron activation at the University of Texas at Austin Nuclear Engineering Teaching Lab's TRIGA MARK II Reactor. The samples were then measured using an ARSA-style beta-gamma coincidence detector. Xe-125 and Xe-127 are of particular interest for international treaty verification as they can occur as emission from research reactors. The latter isotope has also been considered for calibration, QA/QC and tracer for medical isotope production facilities. Isotopically pure beta-gamma spectra of the non-traditional isotopes will be presented and analyzed. Various potential sources and sensitivities for production of non-traditional radioxenon isotopes are also considered.

Log: 451. **PREPARATION OF NON-RADIOACTIVE GLASSY SURROGATE NUCLEAR EXPLOSION DEBRIS (SNED) LOADED WITH ISOTOPICALLY ALTERED NOBLE GASES.** Liezers, M.(1); Carman, A.J.(1), Eiden, G.C.(1). (1) Pacific Northwest National Laboratory.

The measurement of Xe isotope ratios in nuclear explosion debris can be done in a method that requires very little sample preparation. Fragments of fallout debris samples can be heated with a laser or electrically to release trapped gases that include Xe arising from fission product decay, which after clean up can be analyzed in a mass spectrometer. To demonstrate and assess the efficacy of such techniques, we have been investigating a method to incorporate isotopically altered Xe into glass substrate materials that in many respects mimic nuclear explosion debris. Enriched Xe-129 is relatively cheap and fairly widely available as it is used in magnetic resonance imaging. The approach used to prepare these materials will be described along with some of the example results obtained.

Log: 452. **BULK CHEMICAL AND ISOTOPIC VARIATION PROVIDES INSIGHT INTO FALLOUT FORMATION MECHANISMS.** Eppich, G.R. (1); Knight, K.B. (1); Wimpenny, J. (1); Leever, M.E. (1); Spriggs, G.D. (1); Hutcheon, I.D. (1). (1) Lawrence Livermore National Laboratory.

Fallout glass, a byproduct of near-surface nuclear detonations, may be of forensic value in the aftermath of a nuclear event. However, the physical and chemical mechanisms controlling fallout formation remain poorly understood. Here we present major element, trace element, and actinide concentration and isotopic composition data from a suite of aerodynamically-shaped pieces of fallout glass of varying size and morphology from a near-surface nuclear detonation, and we use these data to constrain the formation processes attending fallout formation. Our previous work demonstrated that the bulk chemical composition of fallout resembles the composition of soils from near the site of the nuclear event, while the isotopic composition of uranium in these fallout glasses can be explained by simple two-component mixing between device-derived U ( $^{235}\text{U}/^{238}\text{U} > 8$ ) and natural uranium ( $^{235}\text{U}/^{238}\text{U} = 7.253\text{E-}3$ ). Enrichment and depletion of some elements relative to nearby soils, however, is indicative of chemical fractionation processes, possibly including loss through volatilization and gain through condensation. New data presented here show significant chemical variation with respect to fallout size and morphology. Such behavior suggests incomplete homogenization of the melt produced immediately after detonation, a process halted by rapid quenching. These findings are support a complex formational mechanism involving melting of surrounding surface materials, incorporation of device materials (e.g. highly-enriched uranium) into the melt, loss of volatile elements, mixing and homogenization, agglomeration of discrete molten droplets, and quenching. Prepared by LLNL under Contract DE-AC52-07NA27344. LLNL-ABS-665033.

Log: 453. **K-SHELL FLOURESCENCE YEILDS AND THEIR UNCERTAINTIES FOR USE IN HYBRID K-EDGE DENSITOMETRY.** Nicholson, A.D.(1); Croft, S.(1); McElroy R.D.(1). (1) Oak Ridge National Laboratory.

Hybrid K-edge Densitometry (HKED) is a near absolute, high accuracy, non-destructive analytical assay technique that is used by international nuclear safeguards inspectors to provide rapid determination of accountancy tank solutions. The uranium concentration of spent reactor fuel solutions sampled from the dissolver and the minority element concentrations (e.g. Pu) are determined relative to the uranium concentration using a simultaneous recording of the characteristic K-series fluorescence x-ray spectrum. The physical model used to describe the xray generation process requires that the relative fluorescence yields of the K-shell radiative transitions is known. How to estimate this quantity and how to place a defensible uncertainty on it is the subject of this work. Limited experimental data for the fluorescence yield,  $\omega_K(Z)$ , as a function of atomic number, Z, exists and the data is subject to experimental uncertainty. Current practice is to make use of the available experimental data by performing global fits using a semi-empirical formula. In this work we show the standard form is nonphysical in the actinide region of interest. To describe the variation of  $\omega_K(Z)$ , especially in the actinide region, we use modified expressions and published theoretical values. We use Monte Carlo methods to generate uncertainty estimates, by varying the input experimental data in accordance with the experimenter's assigned uncertainties. The Monte Carlo technique is also used to estimate uncertainties on the relative fluorescence yields,  $(\omega_K(Z))/(\omega_K(92))$ , for the actinides.

Log: 454. **Evaluation of radioisotope labeled emodin for Breast Cancer Diagnosis.** Park, J.H.; Lee, J.Y.; Yang, S.D. Radiation Instrumentation Research Division, Korea Atomic Energy Research Institute.

Emodin is a natural chemotherapeutic compound with diverse biological properties including an antitumor activity. Recently, it has been proven to inhibit HER-2/neu tyrosine kinase expressing breast cancer cells as an anticancer agent. radioisotope [123I] labelled emodin has been synthesized and evaluated on breast cancer which express basal and high levels of HER-2/neu tyrosine kinase to investigate its suitability as a breast cancer diagnosis. The labeling of 123I was performed in [123I]NaI solution(159MBq/100 $\mu$ L), 0.5 M H<sub>3</sub>PO<sub>4</sub>,32% peracetic acid with emodin in ethanol (1mg/mL) then stirring for 10 minutes. Purification of [123I] emodin were carried out by a RP-HPLC with gamma counter using ammonium acetate. The analysis of emodin as a reference were confirmed by NMR, MS spectrometer. The breast cancer cell (SK-BR-3 and MCF7) were used in cell uptake experiments. SK-BR-3 has more expressed HER-2/neu than MCF-7. Cell lines were seeded at in 24 well plates at 1mL per well and incubated at 37°C in a 5% CO<sub>2</sub> for 24h. To each well in [123I]emodin (235 KBq) was added. Cells were washed twice in cold phosphate buffered saline (PBS) at designated time point (10 min, 30 min, 60 min and 120 min). The cell pellets were counted by gamma counter. Data was presented as a percentage of the injected radioactivity dose (%ID). The cellular uptakes of [123I]emodin increased in a time dependent manner for both breast cancer cell lines (SK-BR-3 and MCF7). The uptake of SK-BR-3 was higher than that of MCF-7. The ratio of SK-BR-3 and MCF7 was about 1.7 respectively. In conclusion, we have synthesized and evaluated [123I]emodin as inhibitor of HER-2/neu receptor. [123I]emodin is more effective blocking of SK-BR-3 than MCF-7. It has proven that [123I]emodin has a specific activity on HER-2/neu overexpressed cell. This study suggests [123I]emodin is potent breast cancer imaging reagent.

Log: 455. **ACTINIDES AND 129-IODINE ANALYSIS BY AMS ON A NEW 1MV SYSTEM.** Hotchkis, M.A.C.(1); Child, D.P.(1); Levchenko, V.(1); Wilcken, K.M.(1); Kitchen, R.(2). (1) ANSTO, Sydney, NSW, Australia. (1) National Electrostatics Corporation, Middleton, WI, USA.

In 2014 a new 1MV AMS system has been commissioned at ANSTO. The system is designed to cover the full mass range, with terminal voltage suitable for achieving optimum beam transmission for a wide range of species. In addition to routine radiocarbon analysis at high throughput, this system is planned to be used for analysis of heavy radionuclides, in particular uranium, plutonium and 129-iodine analysis. Up to eight isotopes can be measured concurrently, in an ionisation chamber, Faraday cup or dynode detector. The system has high efficiency and the detection limit for actinide isotopes is below 1 femtogram. The new system is being applied to a range of studies aimed at improving understanding of how actinides behave in the environment. Former nuclear sites, which contain a range of levels of contamination with actinides, offer opportunities to study the migration and bioaccumulation of actinides. In addition to the evaluation of the radiological risk posed to potential human and non-human occupants of those specific sites, such studies can contribute fundamental data to the understanding of the behaviour of actinides in the environment. Performance data and recent results for actinides and 129-iodine will be presented.

Log: 456. **APPLICATION OF VARIABLE SOIL GAS RADON CONCENTRATION MEASUREMENT TECHNIQUES FOR THE PURPOSE OF ENVIRONMENTAL STUDIES.** Fronka, A.(1); Jilek, K.(1); Thinova, L.(2). (1) National Radiation Protection Institute, Prague, Czech Republic. (2) Czech Technical University in Prague, Faculty of Nuclear Sciences and Physical Engineering, Czech Republic.

The first application of soil gas radon concentration measurement that will be presented is related to the radiological protection. The naturally occurring radioactive gas radon (radon-222) is a dominant source of exposure of individuals to ionizing radiation and a major contributor to the annual effective dose assessment. Detailed knowledge of radon transport mechanisms from the subsoil into the indoor environment is essential for the proper design of radon corrective measures. The radon transport behavior is closely associated with the physical properties of building components and materials and the appropriate part of subsoil. Radon transfer factor time variations have been studied based on simultaneous continuous indoor and soil gas radon measurements within the framework of complex radon diagnosis of individual buildings. Independent analysis of radon entry pathways using a specific tracer gas application (N<sub>2</sub>O) was carried out to confirm a new radon transport mechanism from the subsoil underneath the external thermo insulation layer of the building envelope into the indoors. Another application of soil gas radon measurement is linked to the geophysical environmental studies focused on hidden faults confirmation using simple and robust measuring tools that can be applied in demanding climatic field conditions. Variable measuring techniques and methods, including the continuous radon monitors and spot sampling procedures, will be presented and discussed in detail. The overview of geophysical methods applied to above given area of research will be introduced and described in contribution presented by Dr.L. Thinova. The unique application has been recently elaborated in the context of detection of the subsurface radioactive noble gases as a critical component of the on-site inspection (OSI) regime under the Comprehensive Nuclear Test Ban Treaty (Carrigan, Ch., Pure Appl. Geophys. 171, 2014). It appears that existing radon soil gas detection technology can serve as the basis for developing a rapid field survey capability using a near-surface probe sampling system to locate zones of anomalous radon concentration. Radon gas detection systems can be effectively used as an indicator for suitable sampling location of radioactive noble gases generated by an underground nuclear explosion and can be also utilized as the quality control tool indicating a potential leakage of sampling apparatus.

Log: 457. **NRPI MULTI-PURPOSE ON-LINE MONITORING STATION FOR MEASUREMENT OF NATURAL RADIOACTIVITY IN THE AMBIENT ATMOSPHERE AND IN THE SOIL.** Jilek, K.(1); Fronka, A.(1); Hulka, J.(1); Giammanco, S.(2); Neznal, M.(3); Thomas, J.(1). (1) National Radiation Protection Institute, Prague, Czech Republic. (2) Istituto Nazionale di Geofisica e Vulcanologia , Osservatorio Etneo - Sezione di Catania, Italy (3) DIAMO - State Enterprise, Stráž nad Ralskem, Czech Republic.

An automated, on-line and wireless outdoor measurement station of atmospheric radon, gamma dose rate and meteorological parameters was developed and tested at the National Radiation Protection Institute (NRPI) of Prague during the years 2010-2012. The results concerning investigated seasonal and diurnal variations of the outdoor radon concentration have been already published. At the turn of years 2013-2014 the technological enhancement and upgrade of the existing station was completed, regarding the possibility of simultaneous continuous monitoring both of atmospheric radon/thoron decay products and soil gas radon concentration, including measurement of its exhalation rate from the surface soil layers. After introduction of the most important detection, operational and transmission characteristics of the station, which can be a part of network stations, the most interesting results of continuous measurement from the network of stations located within the premises of the NRPI, the DIAMO - state enterprise near above-ground disposal sites of radioactive waste and of the Osservatorio Etneo in an area of Mt. Etna volcano located on a major active fault and exposed to the summit craters gas emissions will be presented and discussed.

Log: 458. **RELATIONSHIP BETWEEN RADON-222 CONCENTRATIONS IN COASTAL WATER AND ENVIRONMENTAL CONDITIONS IN JAPAN.** Honda, H.(1); Taniguchi, M.(1); Ono, M.(2), Hosono, T.(3); Umezawa, Y.(4); Sugimoto, R.(5); Yamada, M.(1). (1) Research Institute for Humanity and Nature. (2) National Institute of Advanced Industrial Science and Technology. (3) Kumamoto University. (4) Nagasaki University. (5) Fukui Prefectural University.

In recent years, submarine groundwater discharge (SGD) have received considerable attention in hydrology and oceanography. Radon-222 ( $^{222}\text{Rn}$ ) is a useful tracer to detect SGD because  $^{222}\text{Rn}$  in groundwater has extremely high concentration compared with surface waters. In Japanese coasts, many researches have been done SGD observations using  $^{222}\text{Rn}$  tracer techniques. If we compiled  $^{222}\text{Rn}$  data from many coasts with different environmental condition, we can get generality related environmental parameters to SGD. Therefore, objectives of this study are to compile the existing observed data in Japanese coasts and to evaluate environmental parameters concerning the regional difference in  $^{222}\text{Rn}$  activities. Firstly, we consolidated  $^{222}\text{Rn}$  data in coastal waters at 10 sites from the northern part to the southern part of Japan. We also prepared environmental parameters in each watershed such as precipitation, geological data and geomorphological characteristics to examine and effect of driving forces on the regional difference of  $^{222}\text{Rn}$  activity. In this presentation, we will show the regional difference of  $^{222}\text{Rn}$  activities in Japanese coasts and statistical analysis results of relationship between compiled  $^{222}\text{Rn}$  data and environmental parameters.



Log: 459. **FIELDABLE MASS SPECTROMETER NEEDS FOR FORENSICS AND SAFEGUARDS.** Dixon, E.T., US Department of Energy, National Nuclear Security Administration, Office of Defense Nuclear Nonproliferation.

The Department of Energy, National Nuclear Security Administration's (NNSA) Office of Defense Nuclear Nonproliferation Research & Development (DNN R&D) supports R&D at the US National Laboratories and their affiliated university and industrial collaborators. Advancements are needed in portable field analytical systems to improve on the timeliness of reporting for nuclear forensics and international safeguards. Existing field analytical capabilities lack the sensitivity, simplicity, speed, and above all, portability, required for routine application. The fielded instrument capabilities of interest primarily include measurements of the isotopic ratios of select actinides and fission products (e.g., Pu, U, Am etc), and secondarily include the compositions of high explosives used in nuclear weapons. The abundance of signature material in a sample may be very low, on the order of 1E-14 grams or less. Principle challenges include: identifying nuclear signatures of interest in a host of environmental backgrounds and matrices; isolating those signatures; and measuring their elemental and isotopic ratios within a total timeframe of <2 hours per sample (or standard) to yield results that would enable the analyst to readily identify nuclear activities. Some current innovations for field applications include excitation using micro-plasmas and miniature lasers, new detector types, new spectrometer types and unique approaches for standardization. However, some of these approaches are too complex or unreliable for routine field application. The integration of reliable components into a carry-on-luggage sized system would enable routine use in forensics and safeguards applications.

Log: 460. **HIGH FLUX NEUTRON GENERATOR FOR NEUTRON ACTIVATION ANALYSIS.** Allan X. Chen (1); Jaakko H. Vainionpaa (2); Melvin A. Piestrup(1), Charles K. Gary (1); Greg Smith (1); David W. Williams (1); David Diprete (3); Glenn Jones (4); Richard H. Pantell (5). (1) Adelphi Technology (2) JHV Physics (3) Savannah River National Laboratory (4) G&J Jones Enterprise (5) Department of Electrical Engineering, Stanford University.

The new model DD110MB compact neutron generator manufactured by Adelphi Technology produces thermal neutron flux that is commonly achievable only by small research nuclear reactors or larger accelerator based systems. The maximum thermal neutron flux at the sample ports has been measured to exceed  $4E7$  n/cm<sup>2</sup>/s. The DD110MB utilizes the deuteron-deuteron (D-D) reaction to produce 2.45MeV fast neutrons by bombarding a titanium target with deuteron at energies up to 120keV. Four microwave ECR ion sources are used to produce a high atomic species of deuterium ions which are then accelerated by a single gap acceleration structure to a central Ti/Cu clad target. An integrated plastic moderator serves to both moderate the fast neutrons and isolate the high voltage target from ground potential. Depending on the choice of the moderator material, the DD110MB can produce a various neutron energy spectrums that are custom-tailored to the desired applications. The total fast neutron yield of the resulting from the D-D reaction exceeds  $2E10$  n/s while using less than 12kW of total beam power. Footprint of the completely shielded DD110MB measures approximately 3m x 3m x 2m, which is ideal for operating inside a modest-sized laboratory room. In contrast to research nuclear reactors and large accelerator systems, the DD110MB presents less administrative and regulatory requirements. We will present both simulation and experimental results that are used to characterize the performance of the neutron generator at the conference.

Log: 461. **UPTAKE OF PLUTONIUM ON A THIN FILM FOR USE IN THERMAL IONIZATION MASS SPECTROMETRY.** David Locklair (1), Joseph Mannion (2), Scott Husson (2), Brian Powell (1). (1) Environmental Engineering and Earth Sciences, Clemson University. (2) Chemical and Biomolecular Engineering, Clemson University. Trace elemental and isotopic analysis of actinides via Thermal Ionization Mass Spectrometry (TIMS) is often difficult and time consuming due to intensive sample preparation. In this work, a polymer thin film ( $\sim 150\text{ }\mu\text{m}$ ) is coated onto a silicon substrate and utilized for rapid radioanalytical analysis. The polymer is comprised of polyvinyl benzyl chloride and functionalized with triethyl amine (TEA) to produce an anion exchange site for concentrating anionic actinide complexes (i.e.  $\text{PuCl}_6^{2-}$ ,  $\text{Pu}(\text{NO}_3)_6^{2-}$ ) from solution. In addition, selectively functionalizing "spots" with TEA creates hydrophilic regions and allows for concentration of an aqueous drop when surrounded by the hydrophobic polymeric backbone. Batch uptake studies were performed using Inductively Coupled Plasma Mass Spectrometry and Alpha Spectrometry to determine uptake and binding capacities of the polymer thin film. Results indicated that along with a potential for utilization as a TIMS substrate, the polymer thin film yields high resolution alpha spectra with sharp peaks, comparable to samples produced via electrodeposition. The thin film geometry allows for rapid Pu uptake kinetics. An equilibrium constant ( $K_d$ ) for the functionalized polymer was found to be approximately 7000. The anion exchange capacity of the film was determined using  $^{36}\text{Cl}$  uptake studies and found to be approximately  $\sim 0.1\text{ meq/g}$ . Thus, the rapid uptake kinetics, high binding capacity, and high resolution alpha spectra show good promise for the use of this thin film for rapid radioanalytical analyses.

Log: 462. **EXTRACTIVE SCINTILLATING RESIN FOR ULTRA-TRACE-LEVEL QUANTIFICATION OF ALPHA- AND BETA- EMITTING RADIONUCLIDES IN ENVIRONMENTAL WATERS.** Duval, C.E. (1); Seliman, A.F. (2); DeVol, T.A. (2); Husson, S.M. (1). (1) Department of Chemical and Biomolecular Engineering, Clemson University. (2) Department of Environmental Engineering and Earth Sciences, Clemson University.

Traditional methods of quantifying alpha- and beta-emitting radionuclides in environmental water samples involve time-intensive, batch style analytical techniques that utilize non-portable instruments. A recent development in environmental sensing is a portable, flow cell detector that utilizes extractive scintillating resin. The extractive scintillating resin serves the dual purpose of (1) concentrating the radionuclide of interest and (2) serving as a radiation transducer. Currently, such resins are produced by physically absorbing organic extractants and fluors into a polymer matrix. Unfortunately, this approach yields resins with poor stability as the active components leach from the resin over time. This contribution describes our work to synthesize a new class of extractive scintillating resin in which the fluor and the extractive ligand are bound covalently within the polymer matrix. Suspension polymerization was used to prepare resin beads comprising 4-vinyltoluene, divinylbenzene, 4-vinylbenzyl chloride, and the fluor, 2-(1-naphthyl)-5-phenyloxazole. The expanded gel resin was functionalized with either mono- or bisphosphonic acid ligands using solid phase synthesis. The resin was characterized by Fourier-transform infrared spectroscopy to support functionalization, and potentiometric measurements to quantify accessible binding sites. Fluorescence properties were studied with confocal laser scanning microscopy to observe the distribution of the fluor, and spectrofluorometry to measure emission wavelength and intensity. Static binding capacity experiments were performed for both resins at pH values above and below the pKa1 and pKa2 of the phosphonic acid groups. Finally, the resin was loaded into the flow cell detector to test the dynamic binding capacity and detection efficiency for uranium.

Log: 463. **Fundamental Solvent Characteristics of ALSEP for Nuclear Fuel Reprocessing.** Brown, M.A.(1); Gullekson, B.J.(2); Kropf, J.A.(1); Paulenova, A.(2); Gelis, A.V.(1). (1) Argonne National Laboratory - Chemical Sciences & Engineering. (2) Oregon State University - Nuclear Engineering and Radiation Health Physics.

Separating the trivalent minor actinides (An) from the lanthanides (Ln) is an important challenge in closing the nuclear fuel cycle in the U.S., particularly when considering long-term geologic repositories. Solvent extraction that uses aliphatic-soluble extractants and an acidic, aqueous, complexing matrix has been the leading technology in reprocessing used nuclear fuel. The acidic extractants HDEHP (di-(2-ethylhexyl)phosphoric acid), HEH[EHP] (2-ethylhexylphosphonic acid mono-2-ethylhexyl ester) and the neutral DGA extractants TODGA (N,N,N',N'-tetraoctyldiglycolamide), and T2EHDGA (N,N,N',N'-tetra(2-ethylhexyl)diglycolamide) can be used to extract and separate Ln/An. When combined, some of these solvents demonstrate adduct formations and synergetic extraction behaviors that can complicate Ln/An separations. These solvent interactions, however, are difficult to characterize and predict. Here we report tracer extraction studies, UV-Visible, FT-IR, and XAF spectroscopies of selected f-elements with these solvents. The results probed the fundamental thermodynamic and structural aspects of the Ln/An-solvent complexes and helped characterize which solvents form adducts. While these interactions are still under investigation, the interpretations of the results presented here bring the ALSEP processes closer to a complete fundamental understanding and closer to larger-scale pilot demonstrations.

Log: 464. **RADIOCHEMISTRY SEPARATIONS WITH SILICA-GRAFTED CMPO-MODIFIED CALIX[4]ARENES.** May, E.(1); Solovyov A.(2); Matveev Y.(3); Guo, Y.(2); Kalchenko, V.(3); Katz, A.(2); Nitsche, H. (1,4) (1) Department of Chemistry, University of California, Berkeley. (2) Department of Chemical and Biomolecular Engineering, University of California, Berkeley. (3) Institute of Organic Chemistry, National Academy of Sciences of Ukraine, Kyiv, Ukraine (4) Lawrence Berkeley National Laboratory, Berkeley, CA.

Since their discovery, calixarenes, whether modified or unmodified, have proven useful for a diverse range of elemental separations within many different fields of chemistry. Of particular interest is their use in radiochemistry, where ligands relevant to nuclear reprocessing, such as carbamoylmethylphosphine oxide (CMPO), have been attached to the upper or lower rims of calixarenes and the separation behavior compared to that of the free ligand. In an overwhelming majority of these cases, radionuclide separation was significantly enhanced by using the calixarene to constrain the ligands in a favorable manner. Most of these studies have been performed in the context of liquid-liquid separations and have involved modification to the calixarene for solubility purposes. In this study, the CMPO-modified calix[4]arene was anchored, via the lower rim, to the internal surface of a porous silica support. The overarching goal was to investigate the influence of the anchoring approach (grafting versus tethering) to the surface of silica, and comparing these two anchoring methods with CMPO-calixarene liquid-liquid extraction. The aim of such a comparison is to show the importance of surface interactions in the anchored site, which may enhance performance in a manner that is impossible to achieve when using soluble ligands in solution.

Log: 465. **ESSENTIAL ELEMENT CONTENTS IN FOOD GROUPS FROM THE SECOND BRAZILIAN TOTAL DIET STUDY.** Ambrógi, J.B; Avegliano, R.P.; Maihara, V. A. Instituto de Pesquisas Energéticas e Nucleares- IPEN - CNEN/SP, São Paulo, Brazil.

Since 1978 Total Diet Studies (TDS) have been considered as the most appropriate approach to estimate dietary exposure of contaminant and essential elements by the World Health Organization. This paper presents results of the average dietary estimations of Ca, Cr, Co, Fe, K, Na, Se and Zn from the second Brazilian Total Diet Study (TDS). The food consumption data source used was from the 2008-2009 Household Budget Survey (HBS) by the Brazilian Institute for Geography and Statistics. Food list was composed by a total of 82 food items which were grouped into 19 food groups according to the similar nutritional composition. The food groups were: Cereals; Beans; Vegetables; Fruits; Nuts and seeds; Flours, pasta and bread; Cakes and biscuits; Cattle meat; Pork; Poultry; Fish; Industrialized meat and offal; Eggs; Dairy products; Sweets; Oils and fats; Beverages; Pizza, snacks, sandwiches, soups, sauces, mixtures; Salt. This Food List reflects 100% of the daily individual food consumption inside and outside of the households of the Brazilian southeastern population. Instrumental Neutron Activation Analysis was used to determine the concentration of the essential elements (Ca, Cr, Co, Fe, K, Na, Se and Zn). A large variation among food groups were observed for the average concentration of the most elements. The dietary intake values were compared to the 1st Brazilian TDS that used data from 2002-2003 HBS.

Log: 466. **RADIOLOGICAL ASSESSMENT OF PHARMACEUTICAL CLAYS.** Silva, P.S.C.(1); Máduar, M.F.(1); Scapim, M.A.(1); Garcia, R.H.L.(1); Martins, J.P.M.(1). (1) Instituto de Pesquisas Energéticas e Nucleares.

Clay minerals have been used for therapeutic purposes since ancient time in wound healing, skin irritation relieve, as anti-inflammatory and to treat gastrointestinal disorders. Currently, it is also highlighted its use in the pharmaceutical industry as active ingredient, due to its high absorption capacity, specific surface and easy handling as excipient. Clays also are used to promote drug disintegration, influencing the processes of medicine liberation when orally administered. Furthermore, they are used as cosmetics owing to its capacity of fats and toxins adsorption. Although, it is considered as a natural material, clays may not be free of possible adverse health effects due to the presence of hazardous minerals, radioactive and toxic elements. In this study eight raw clay samples and six commercial clay samples were analyzed to determine its mineralogical, chemical and radiological characteristics as well as its suitability for medicinal and cosmetic applications. The mineralogy of the sample was determined by X-Ray diffraction, elemental concentration was determined by X-ray fluorescence and neutron activation analysis and the activity concentrations of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{40}\text{K}$  were determined by gamma spectrometry. For radiological characterization the absorbed dose rate, gamma representative level index, excess lifetime cancer risk, external radiation hazard and internal radiation hazard indices as well as the absorbed dose due to ingestion were determined. Results indicated that some clays may be enriched in elements such as As, Br, Cr and Zn. Mean activity concentrations were 424 Bq/kg, 51,2 Bq/kg, 41,5 Bq/kg and 44,5 Bq/kg for  $^{40}\text{K}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  respectively.



Log: 467. **INVESTIGATION OF THE RELEASE AND MOBILITY OF AMERICIUM FROM REFRACTORY PLUTONIUM AT THE SITE OF THE BOMARC ACCIDENT.**

Gallardo, A.M.(1); Sudowe, R. (1). (1) University of Nevada Las Vegas.

The Boeing Michigan Aeronautical Research Center (BOMARC) operated as a nuclear missile defense site from 1958-1971. In June of 1960, a missile carrying a nuclear warhead was partially consumed by a fire which resulted in the release of weapons grade plutonium (WGP). The WGP was scattered over a 7-acre area due to turbulent local atmospheric conditions and the water applied as part of the fire extinguishing measures. The WGP hot particles in the BOMARC soil exist in the form of high-fired PuO<sub>2</sub>. These refractory particles consist of a matrix containing <sup>239</sup>Pu, <sup>240</sup>Pu and <sup>241</sup>Am. The americium is expected to be trapped in the refractory matrix together with the plutonium; however several studies have found the presence of <sup>241</sup>Am outside of this matrix. This research is part of a study that will aid in the investigation of the mechanisms that resulted in the release and migration of <sup>241</sup>Am from the refractory matrix. A sequential extraction technique was used to investigate the association of these actinides with specific host phases in the BOMARC soil. Sequential extraction involves the leaching of elements with a succession of increasingly aggressive chemical treatments that attack specific environmental conditions. The radioisotopes from each leachate were separated using extraction chromatography and prepared for alpha spectroscopy using cerium fluoride microprecipitation. The results of this study can be used to model responses for future situations involving the accidental or intentional release of WGP.

Log: 468. **DETERMINATION OF RADIOISOTOPES IN COMPLEX SALINE MATRICES USING EXTRACTION CHROMATOGRAPHY AND LIQUID SCINTILLATION COUNTING.** Daum, J.K.(1); Sudowe, R.(1). (1) University of Nevada Las Vegas.

Nuclear operations conducted over the past seventy years have resulted in the contamination of the environment with potentially hazardous radioactive species. Atmospheric testing of nuclear weapons along with accidents such as the ones that occurred in Chernobyl, Ukraine and Fukushima, Japan are some of the major sources of anthropogenic radionuclides in the environment. Sea and ocean water cover more than two-thirds of the Earth's surface and are therefore the major recipients of radioactive contamination in the environment through either atmospheric or aquatic pathways. It is therefore critical to conduct research into the separation and rapid determination of radioactive elements from highly saline matrices. A number of radioanalytical techniques are commonly used to determine radioisotopes within a variety of matrices. However, the elevated salinity of sea and ocean water imparts a complex matrix that could affect the isolation, characterization, and determination of the radionuclides of interest. Modern radioanalytical separation techniques often employ extraction chromatographic resins in order to separate radionuclides from sample matrices. Batch experiments and column studies were conducted in order to determine the retention capabilities of the extraction chromatographic resins for elements of interest, particularly isotopes of plutonium and americium, within an artificial ocean water matrix. The selective extraction of the radioisotope was assayed by liquid scintillation counting (LSC) to determine the retention capability of the resin. The retention capabilities of the resins were measured by determining the capacity factor,  $k'$ , which is the free column volumes to peak maximum.

Log: 469. **NEW PROMPT GAMMA ACTIVATION ANALYSIS CAPABILITY FOR THE ASSESSMENT OF SILICON IN BIOLOGICAL MATERIALS.** Maria I. V. Martinez (1,2), Dağıstan Şahin (1), Rolf Zeisler (1), Elisabete A. De Nadai Fernandes (2), (1) National Institute of Standards and Technology (NIST), USA, (2) Centro de Energia Nuclear na Agricultura (CENA), Brazil.

A new cold neutron prompt gamma activation analysis (CNPAA) instrument has come online at the NIST Center for Neutron Research (NCNR). Its key features are a significantly improved neutron flux combined with greatly reduced background when compared to the existing CNPAA instrument at the NCNR. Particularly valuable is the reduction in Si background from 3900  $\mu\text{g}$  to about 40  $\mu\text{g}$ , which is acceptable for the determination of the element in plant materials. In the vast majority of plants species, Si is not considered an essential nutrient but a "quasi-essential" element, which improves the plant health and provides increased resistance to pest, disease and hydric stress. The current study includes a candidate sugarcane reference material developed by CENA with an expected nutritionally adequate Si mass fraction level of 0.5 % to 0.75 %. As matrix dependent flux depression through neutron scattering is observed in CNPAA, methods of correction for the accurate determination of Si are developed. The performance of the instrument is evaluated with a variety of plant reference materials and the sugarcane candidate reference material.

Log: 470. **Separation of  $^{163}\text{Er}$  from alpha particle irradiated dysprosium target: A step toward neutrino mass measurement through electron capture of  $^{163}\text{Ho}$ .** Moumita Maiti, Zoltan Szucs, Susanta Lahiri.

Among the different approaches for neutrino mass measurements, experiments exploring electron capture processes of suitable nuclides can provide necessary information on the electron neutrino mass value.  $^{163}\text{Ho}$  is a candidate radionuclide due to its low Q-value of 2.8 keV for the electron capture. Earlier  $^{163}\text{Ho}$  was produced by proton-spallation reaction on Ta target, or by neutron activation using  $^{162}\text{Er}(n,\gamma)^{163}\text{Er}(\text{EC})^{163}\text{Ho}$ . Proton induced reaction on natural dysprosium target would also lead to the production of  $^{163}\text{Ho}$ .  $^{163}\text{Ho}$  (4570 yr) has no alpha, beta or gamma radiation, therefore cannot be monitored during its separation from dysprosium. We proposed for the first time two indirect production routes of  $^{163}\text{Ho}$  in LTD-14 conference, 2011, in Heidelberg. These two routes are (i)  $\text{Dy}(\alpha, xn)^{163}\text{Er}(\text{EC})^{163}\text{Ho}$  (ii)  $\text{Tb}(7\text{-Li}, 3n)^{163}\text{Er}(\text{EC})^{163}\text{Ho}$ . Advantages of these methods are  $^{163}\text{Er}$  has only 75 min half-life and separation of Dy-Er may be easier than Dy-Ho pair. We therefore irradiated natural dysprosium oxide target at the Variable Energy Cyclotron Centre, Kolkata, India with 40 MeV alpha-particles followed by separation of Er radionuclides from dysprosium target by liquid-liquid extraction technique using di-(2-ethylhexyl) phosphoric acid (HDEHP) dissolved in cyclohexane as organic reagent. The irradiated dysprosium target was dissolved in HCl and extracted with 1% HDEHP and 0.2 M HCl. About 84% Er and 49% Dy was extracted in the organic phase, which was again back extracted by 0.5 M HCl. The back extracted aqueous phase contained about 49% Er with less than 1% dysprosium contamination. Proposed chemical method is fast enough to complete the entire chemical process within one half-life of  $^{163}\text{Er}$ .

Log: 471. **AGRICULTURAL MANAGEMENT, SEASON AND TRACE ELEMENTS EFFECTS ON VOLATILE OIL PRODUCTION FROM MELISSA OFFICINALIS L. (LEMON BALM).** Sussa, F.V.; Duarte, C.L.; Silva, P.S.C. Instituto de Pesquisas Energéticas e Nucleares.

In recent decades, the study of medicinal plants has become the focus of ever more extensive research all over the world, due to diversity and potential that medicinal plants have as source of medicinal products. The *Melissa officinalis* medicinal properties are related to the wide variety of its chemical composition volatile oil, such as terpenoids. These are a large group of secondary metabolites, which can vary qualitatively and quantitatively, depending on various factors such as fertilization, season and nutrient availability in the soil. Providing information on agricultural management, season and trace elements effects on volatile oil is fundamental to obtain the highest production by the specie. *Melissa officinalis* is a medicinal and aromatic species popularly used to promote sleep, reduce stress and anxiety. Their medicinal properties are related to the major compounds of their volatile oil, just like the citronellal, neral and geranial. Container experiments were carried out in the Municipal Gardening School experimental area, in Ibirapuera Park, São Paulo, Brazil. The experimental design was completely randomized with three treatments: control (site soil), organic fertilization (2t/ha of poultry manure) and conventional fertilization (30t/ha of NPK, 6:14:8) in four distinct seasons (spring, summer, autumn and winter). Elemental concentration for the elements As, Ba, Br, Ce, Cl, Co, Cr, Cs, Eu, Fe, Hf, K, La, Lu, Mg, Mn, Na, Nd, Rb, Sb, Sc, Se, Sm, Ta, Tb, Ti, Th, U, V, Yb, Zn and Zr was determined by Instrumental Neutron Activation Analysis (INAA) in *Melissa officinalis* leaves. The analysis of elements Cd, Pb, Ni and Cu was performed using atomic absorption spectrometry (AAS). Certified reference materials IAEA-336 and NIST SRM-1573a Tomato Leaves were analyzed for quality control. The volatile oil was extracted from its leaves by hydrodistillation process in Clevenger apparatus and analyzed by gas chromatography coupled to a mass spectrometer (GC-MS). The major compounds neral, geranial and citronellal were identified by using reference standards from Sigma Aldrich. Data analysis was done by chemometrics such as Analysis of Variance (ANOVA), Correlation Analysis (CA), Hierarchical Cluster Analysis (HCA) and Principal Component Analysis (PCA).

Log: 472. **QUANTITATIVE EVALUATION OF MODAL VARIABILITY IN AERODYNAMIC FALLOUT GLASS.** Marks, N.E.(1); Weisz, D.G.(1,2); Jacobsen, B.(1); Knight, K.B.(1); Hutcheon, I.D.(1). (1) Lawrence Livermore National Laboratory; (2) University of California, Berkeley.

Aerodynamic fallout glass forms as a result of near surface nuclear explosions when sediment at the blast site is swept into the fireball and vitrified. The chemical composition of fallout glass represents an admixture of entrained surface sediments and trace quantities of device material, and may include depletions in volatile elements including carbon and sodium. Quantitative X-ray mapping is increasingly used in the geological sciences to investigate textures, mineral zoning, and element distribution in rocks. In this study we combine elemental mapping with quantitative image analysis to constrain the conditions of formation of glassy fallout. We analyzed over a dozen individual glassy objects from a near surface nuclear test to better understand relative chemical abundances, phase relationships, and chemical components of the aerodynamic glassy fallout. The fallout glasses are generally heterogeneous in composition; the objects commonly have two or more end-member compositions ranging from low-Si mafic glass to high-Si rhyolitic compositions. The objects commonly contain relict grains of vitrified quartz (>99% SiO<sub>2</sub> glass), as well as vesicles ranging from <1 to 100s of microns. Compositional banding in some of the objects is interpreted to result from flow of material within the molten objects. The bulk compositions appear similar to background sediments present at the test site. Electron backscatter diffraction analyses indicate that the fallout beads from a uranium-fueled test are entirely glassy, with no relict crystalline phases present. The presence of nearly euhedral, vitrified relict grains with sharp compositional boundaries implies rapid heating and quenching of the glassy fallout beads.

Log: 473. **CHARACTERIZATION OF A NEW REFERENCE MATERIAL – A TRIVIAL TASK WITH INAA?** Maria I. V. Martinez (1,2), Rolf Zeisler (1), Elisabete A. De Nadai Fernandes (2), (1) National Institute of Standards and Technology (NIST), USA, (2) Centro de Energia Nuclear na Agricultura (CENA), Brazil.

The sugarcane production is one of the most important economic activities in Brazil and research in cultivation and nutrition of the plants is a high priority. A candidate reference material of sugarcane leaves had been prepared by CENA in support of analytical measurements. The leaves had been dried, pulverized, sieved, and portioned in brown plastic bottles (410 bottles, 20g each). INAA was selected to determine the homogeneity of the bottled material and to provide quantitative results for trace elements suitable for value assignment. Samples were prepared from 10 randomly selected bottles to determine the dry mass, to assess the homogeneity, and to determine mass fractions of minor and trace elements. INAA with short and long irradiations was performed on duplicate 180 mg test portions as well as one small 2 mg test portion from each bottle. Statistical analysis of the results for the large sample sizes confirmed acceptable representativeness for these test portions. Applying the Kurfürst homogeneity factor model to the data showed acceptable homogeneity of Al, Ca, Mg, and Mn even in the small test portions. However an about 5% bias was discovered in SRM control materials for elements with short-lived nuclides during irradiation using a standard stacked pellet arrangement of filter paper standards and samples. Flux mapping in the rabbit confirmed 5% drops of neutron flux over 1 mm thick pellets of filter paper or biological material when placed perpendicular to the rabbit axis. Placing samples and standards parallel to the rabbit axis allowed for correct results. Further investigations on this neutron beam like effect are underway.

Log: 474. **DEVELOPMENT OF A SUBMERSIBLE RADON SURVEY SYSTEM FOR MAPPING GROUNDWATER DISCHARGE.** Breier, J.A. (1); Peterson, R.N. (2); Bailey, J. (1); Gomez-Ibanez, D. (1); Sayre-McCord, R.T. (1); Singh, H. (1); (1) Woods Hole Oceanographic Institution. (2) Coastal Carolina University.

Sediment pore-waters are often chemically distinct from surface waters. When sediment pore-waters discharge to surface waters, they modify surface water chemistry. Groundwater discharge to the coastal zone and fluid seepage along continental margins are two general examples of this process. This type of exchange can have significant ecological consequences. For instance, nutrient enriched groundwater discharge can impact coral ecosystem function by promoting the growth of benthic macroalgae. This type of exchange can be difficult to detect; but, naturally occurring radon has proven to be a useful tracer of groundwater discharge. Previous studies have employed surface based radon measurement techniques to measure and map dissolved radon gas concentrations for groundwater discharge mapping. However identifying benthic discharge from surface measurements becomes increasingly difficult with depth. Over the past several years, we have been developing a submersible radon measurement system that can be used to extend the utility of this tracer to mapping fluid discharge at deeper depths. This system operates at ambient pressures, equilibrates the carrier gas with ambient radon concentrations by direct sparging, and is being designed for multiple modes of deployment including towed surveys and remote and autonomous vehicle deployments. A prototype system is being used in field trials in Cape Cod, Massachusetts.

Log: 475. **ANALYTICAL RADIOCHEMISTRY ROUTINES AND METHODS FOR PU-238 PRODUCTION DEMONSTRATION AT ORNL.** Delashmitt, J.S. (1); Canaan, R.D. (1); Giaquinto, J.M. (1); Roach, B.D. (1); Smith, R.R. (1); (1) Oak Ridge National Laboratory.

A technology demonstration is currently underway to provide a new supply of plutonium-238 using existing nuclear research reactors at the Oak Ridge National Laboratory (ORNL) and Idaho National Laboratory (INL) and existing chemical processing facilities at ORNL. This demonstration is the first step towards the reestablishment of the domestic production of Pu-238 fuel radioisotope power systems for the National Aeronautics and Space Administration (NASA) space vehicles. Currently at ORNL, target design qualification; target fabrication; irradiation; and chemical processing development tasks have begun to develop appropriate technologies for implementation towards full-scale production. The Nuclear Analytical Chemistry and Isotopics Laboratories (NACIL) Group leads the analytical effort for this project which includes providing the experimental data required for the qualification of the irradiated Np-237 oxide targets, development of analytical methods for the separations flow sheet testing for recovery of plutonium from bulk neptunium dissolver solutions, and the eventual qualification of the final Pu-238 product. A selection of novel analytical techniques will be presented, including radiochemical measurements for Pu-236 content in the Pu-238 product; ion chromatographic (IC) measurements of organic and aqueous hydroxylamine and nitrite levels in the separations flow sheet testing; hot-cell resin separations of trace impurities for analysis by Inductively Coupled Plasma Mass Spectrometry (ICP-MS); and hot-cell auto titration methods for accurate free- and bound-acid determination. This research was sponsored by the U.S. Department of Energy, Office of Nuclear Physics, and funded by NASA. The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-00OR22725. Accordingly, the U.S. Government retains a non-exclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

Log: 476. **Reaction Rate Measurements at the National Criticality Experiments Research Center.** Bredeweg, T.A.(1); Bounds, J.A.(1); Brooks, Jr., G.H.(1); Favorite, J.A.(1); Friese, J.I.(2); Goda, J.M.(1); Greenwood, L.R.(2); Gregory, S.J.(2); Hayes, D.K.(1); Jackman, K.R.(1); Little, R.C.(1); MacInnes, M.R.(1); Metz, L.A.(2); Myers, W.L.(1); Oldham, W.J.(1); Olson, A.C. (1); Rundberg, R.S.(1); Sanchez, R.G.(1); Schake, A.R.(1); White, M.C.(1); Wilkerson, Jr., C.W.(1). (1) Los Alamos National Laboratory, Los Alamos, NM, 87545, USA. (2) Pacific Northwest National Laboratory, Richland, WA 99352, USA.

With the resumption of regular operations of the Los Alamos Critical Assemblies at the National Criticality Experiments Research Center (NCERC), located at the Nevada National Security Site, we have embarked upon a series of campaigns to restore the capability to perform integral reaction rate and fission product yield measurements using historical radiochemical methods. This talk will present an overview of the current and future experimental plans, including results from our experimental campaigns on the Comet/Zeus and Flattop assemblies.



Log: 477. **ADVANCED DRY HEAD-END TREATMENT OF USED NUCLEAR FUEL BY NO<sub>2</sub>**. Johnson, J.A.(1); DelCul, G.D. (1); Collins, E.D.(1); Spencer, B.B.(1); Hunt R.D.(1). (1) Oak Ridge National Laboratory.

The US Department of Energy continues to explore innovative methods of reprocessing used nuclear fuel that have the potential to reduce overall cost and radioactive gaseous emissions. One process being investigated is a dry head-end treatment of the fuel with NO<sub>2</sub> gas before subsequent processing. This process comminutes the mostly monolithic ceramic oxide fuel by further oxidation, producing a finely divided fuel powder and releasing volatile and semi-volatile fission products at temperatures significantly lower than other previously explored dry head-end processes. The product of the initial reactions is  $\epsilon$ -UO<sub>3</sub>, which can be further reacted with NO<sub>2</sub> at reduced temperatures and increased pressure to form a uranyl nitrate salt. In addition to the ability to release volatile and semi-volatile fission products up-front for trapping, the products themselves are attractive for subsequent aqueous processing because of the elimination of massive NO<sub>x</sub> production upon dissolution and the selective solubility properties of nitrated fuel. Numerous facets of the process have been explored with both simulated and irradiated nuclear fuel, including oxidation kinetic rates,  $\epsilon$ -UO<sub>3</sub> structure, iodine removal efficiencies, and oxidant recycle. Results of these studies and a proposed roadmap for further investigation will be presented.

Log: 478. **PRESERVING HIGH-PURITY URANIUM-233.** Krichinsky, A.M. (1); and Giaquinto, J.M. (1); (1) Oak Ridge National Laboratory.

Plans are being made for downblending and disposing of the stockpile of a valuable synthetic isotope of uranium,  $^{233}\text{U}$ , because it has become too onerous for the Department of Energy (DOE) to store indefinitely. Currently, only a small portion of the U.S. holdings of separated  $^{233}\text{U}$  is being kept from downblending with depleted uranium and ultimate disposal (scheduled to begin in 2019). The materials being preserved thus far include only 1 kg of very high isotopic purity  $^{233}\text{U}$  (>99%), mainly intended to replace a diminishing supply of certified reference material (CRM), plus another 44 kg of 97%-pure  $^{233}\text{U}$  to support subcritical experiments. However, many additional kilograms of  $^{233}\text{U}$  (97% to 99% pure) are still destined to be downblended, which will permanently destroy the potential value of this material for many other applications. Furthermore, it is not likely that this material will ever be replaced due to a lack of operating production capability and the high cost of replacing or restarting such capability used in the past. This paper delineates key activities of the preservation process: (1) clean chemistry protocols implemented to ensure isotopic integrity of individual batches; (2) analytical measurements adapted to these high-purity materials; (3) gamma spectrometry measurements with ultra-pure  $^{233}\text{U}$ ; and (4)  $^{229}\text{Th}$  recovery activities. In addition, this paper summarizes  $^{233}\text{U}$  production, use, and ongoing plans to dispose of or to preserve portions of the stockpile. Other identified needs brought forth by the scientific community at the MARC X workshop on this topic also are presented. However, organizations needing  $^{233}\text{U}$  should recognize the importance of making known their interest in preserving other high-purity  $^{233}\text{U}$  materials and ensuring that their needs are conveyed to the DOE. Research is sponsored by the National Nuclear Security Administration. The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-00OR22725. Accordingly, the U.S. Government retains a non-exclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

Log: 479. **FAST NEUTRON DETECTION AND SPECTROSCOPY BY USE OF COMPOSITE CAPTURE-GATED SCINTILLATORS.** Mayer, M.; Nattress, J.; Foster, A.; Barhoumi Meddeb, A.; Trivelpiece, C.; Ounaies, Z.; Jovanovic, I. The Pennsylvania State University.

Fast neutron detection can be accomplished by two principal methods. In the first approach, the scintillation and/or ionization following nuclear recoil produced by the neutron elastic scattering is utilized. In the second approach, the neutron is first thermalized and then captured, whereby energetic charged particle products from neutron capture are detected. Only the first class of approaches allows measurements of the neutron spectrum, which is unfolded using the known, separately measured detector response. Since the unfolding relies on the entire spectrum, neutron energy cannot be measured on an event-by-event basis. A hybrid approach based on measurement of the energies produced by both thermalization and capture circumvents this limitation. To date, capture gated scintillation detectors have relied on employing a thermalization agent that exhibits pulse discrimination properties in order to reject the gammas that usually accompany neutrons. We describe an alternative approach to capture-gated neutron detection that does not employ materials that exhibit pulse shape discrimination, yet it accomplishes excellent neutron/gamma discrimination and should allow direct neutron spectroscopy. By combining scintillating polyvinyl toluene and scintillating  $^6\text{Li}$ -doped glass rods cut into optimal dimensions, the high level of gamma rejection results from both the differences in neutron cross section, partition of charged particle energy deposition between glass and plastic, and differences in scintillation decay times. We review our recent progress on design, development, and measurements employing this composite detector paradigm, which offers significantly greater latitude in detector material selection. Potential benefits for imaging, scientific, industrial, and nuclear security applications will be outlined.

Log: 480. **DOE JAPAN AIR FILTERS: GAMMA SPECTROSCOPY ANALYSIS AND DATA EVALUATION.** Wimer, N.G.; Lawrence Livermore National Laboratory.

The U.S. Dept of Energy field response for the Fukushima Daiichi Nuclear Plant accident in Japan (March 2011) performed air-filter collections varying widely in sampling date, location, and duration. The majority were paired filter papers and charcoal cartridges, in a monitoring configuration standard for the nuclear power plant industry. After initial-screening gamma assays in Japan, all 288 such filter pairs were transported for U.S. fixed laboratory gamma assay, and eventual radiochemistry. Air concentrations of radionuclides are reported, for papers and cartridges gamma-assayed separately. Challenges are described in laboratories achieving needed accuracy and precision in the assay of samples from nuclear power plant accidents. Remarks are made on Fukushima Daiichi source-term composition, relative particulate and gaseous physical forms over time, variation in radionuclide composition with geographic location and altitude, differences between standard nuclear power plant monitoring approaches and the Fukushima accident release composition, the need for better characterization of filter-media trapping efficiencies, and lessons learned for the DOE air filter collection technique. Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract DE-AC52-07NA27344.

Log: 481. **SPECIATION OF TECHNETIUM IN HIGHLY ACIDIC MEDIA (CF<sub>3</sub>SO<sub>3</sub>H, H<sub>2</sub>SO<sub>4</sub>).** Ibtihel Denden (1), Jérôme Roques (2), Frédéric Poineau (3), Massoud Fattahi (4). (1) CEA, DSM, IRAMIS, F-91191 Gif-sur-Yvette, France. (2) Institut de Physique Nucléaire, 91406 ORSAY, France. (3) Department of Chemistry and Biochemistry, University of Nevada, Las Vegas. (4) Subatech Laboratory, 44322 Nantes, France.

This project is part of technetium speciation in highly acidic media. The behavior of Tc in trifluoromethanesulfonic acid was carried out in the absence then in the presence of  $\alpha$  irradiation. Given these two conditions, spectrophotometric results are similar. XAS analysis indicates the formation of a cyclic dimer of Tc(IV) complexed to triflate ligands and formulated as Tc<sub>2</sub>O<sub>2</sub>(CF<sub>3</sub>SO<sub>3</sub>)<sub>4</sub>(H<sub>2</sub>O)<sub>4</sub>. This compound is linearized to TcIV-O-TcIV with the increase of HTFMS concentration. At high concentration of HTFMS +98% (11.15 M), the protonated species TcO<sub>3</sub>(OH)(H<sub>2</sub>O)<sub>2</sub> which is formed in the absence of external ionizing radiations, is reduced to the V oxidation state under  $\alpha$  irradiation. Structural characterization by EXAFS spectroscopy and DFT calculations suggests the formation of monomer species of Tc(V)-triflate complexes where [OTc(F<sub>3</sub>CSO<sub>3</sub>)<sub>2</sub>(H<sub>2</sub>O)<sub>2</sub>]<sup>+</sup> and [OTc(F<sub>3</sub>CSO<sub>3</sub>)<sub>2</sub>(OH)<sub>2</sub>]<sup>-</sup> compounds were proposed. In concentrated H<sub>2</sub>SO<sub>4</sub> (CH<sub>2</sub>SO<sub>4</sub>  $\geq$  12 M),  $\alpha$ -radiolysis experiments of Tc(VII) were performed in order to compare the radiolytic behaviour of Tc(VII) in both comparable media HTFMS and H<sub>2</sub>SO<sub>4</sub>. XANES studies show that radiolytic reduction of Tc(VII) leads to the formation of Tc(V)-Tc(VII) mixture in H<sub>2</sub>SO<sub>4</sub> 13 M and just Tc(V) in 18 M of H<sub>2</sub>SO<sub>4</sub>. The analysis of EXAFS spectra is consistent with the formation of [TcO(HSO<sub>4</sub>)<sub>3</sub>(H<sub>2</sub>O)<sub>2</sub>] and [TcO(HSO<sub>4</sub>)<sub>3</sub>(H<sub>2</sub>O)(OH)]<sup>-</sup> monomer complexes in H<sub>2</sub>SO<sub>4</sub> 13 M and [Tc(HSO<sub>4</sub>)<sub>3</sub>(SO<sub>4</sub>)(H<sub>2</sub>O)] and [Tc(HSO<sub>4</sub>)<sub>3</sub>(SO<sub>4</sub>)(OH)]<sup>-</sup> species at 18 M of H<sub>2</sub>SO<sub>4</sub>.

Log: 482. **MICRODOSIMTERY OF ALPHA-EMITTING DECAY PRODUCTS IN TISSUE USING CONVENTIONAL FILM AUTORADIOGRAPHY.** Schneider, N.R.(1); Spitz, H.B.(1); Glover, S.E.(1). (1) University of Cincinnati.

Previous biodistribution study of the  $^{212}\text{Pb}$ -trastuzumab, a HER2/neu-targeting immunoglobulin (monoclonal antibody), identified that the compound was present in several organs and tissue of the mouse within hours post IV injection by gamma spectrometry. Although the radiolabeled antibody was intended to irradiate prostate tumors with high-energy alpha particles from  $^{212}\text{Po}$ , a short-lived (10.64 hr) decay product of  $^{212}\text{Pb}$ , results of the biodistribution study suggest that other healthy tissues were at risk of irradiation. A study of the microdistribution of alpha emitters in tissues is planned using conventional autoradiographic methods with tissue samples from subjects who were injected with thorotrast, a radiographic contrast medium historically used in diagnostic cerebral angiography or hepatosplenography exams. This work describes a practical method using conventional photographic films and materials for imaging alpha tracks in tissue to determine where in the tissue maximum energy deposition occurs. Conventional photographic films that provide sufficient resolution for autoradiography are becoming difficult to obtain commercially. Although other films can provide a response to alpha energy by exhibiting increased optical density, only films with high resolution are able to produce individual images of alpha tracks that can identify the deposition site of the alpha emitter. This study used Ilford Pan F Plus 135-36, Black & White, ISO 50 film for obtaining images of alpha tracks. The method was developed using a variety of alpha radiation sources including electrodeposited planchets and samples of bone containing  $^{226}\text{Ra}$  that were placed directly on the film for 12 to 48 hours. Post exposure the film was developed using standard darkroom techniques with Kodak D-76 developer for 7 minutes, stopbath, and fixer. Tracks consisting of 5 or more grains were counted using an Olympus BX51 microscope at 40x magnification in a 9.08 cm<sup>2</sup> circular field-of-view. Initial analysis shows an efficiency of approximately 34.9% with an uncertainty in track counts of  $\pm 2.4\%$  for the monoenergetic particles.

Log: 483. **PRESERVING 244Pu IN MARK-18A TARGETS AND THE BENEFIT TO PLUTONIUM CHEMISTRY.** Bradley Patton(1), Sharon Robinson(1), Sheri Bone(2), and Steve LaMont(3).(1) Oak Ridge National Laboratory, (2) U. S. Department of Energy, (3) Los Alamos National Laboratory.

From the 1960's-80's, the Department of Energy (DOE) irradiated targets in high-flux reactors at the Savannah River Site (SRS) to create special long-lived radioactive isotopes for DOE programmatic use, scientific research, and industrial and medical applications. The reactors are now shut down, and it would be prohibitively difficult and expensive to produce these types of heavy isotopes in existing reactors. The 244Pu and heavy curium contained in the Mark-18A targets stored at SRS are examples of these unique materials. The DOE National Strategic Plan for Nuclear Materials (2011) identified these valuable radionuclides as rare and economically irreplaceable, and an Inspector General's audit report (2013) recommended making a decision to preserve the materials before the opportunity is lost. DOE is developing a program to preserve these materials by retrieving the Mark-18A targets from storage at SRS, processing the materials at Savannah River National Laboratory (SRNL) to separate/recover the 244Pu and heavy curium, and transferring them at ORNL for beneficial use. The Mark-18A source of 244Pu has a number of benefits to the plutonium radiochemistry community. Plutonium-244 is an extremely rare and long-lived isotope of plutonium that is not produced in commercial fuel or weapons-grade plutonium, which makes it ideal as certified reference material. Plutonium-244 has a number of other unique and important uses in nuclear forensics and safeguards programs, super heavy element research, and basic plutonium research. This paper provides reviews of programs that would benefit from the use of 244Pu, and provides the purity and quantity needs for each application.

Log: 485. **FUTURE XENON SYSTEM OPERATIONAL PARAMETER OPTIMIZATION.** Lowrey, J.D.(1); Eslinger, P.W.(1); Bowyer, T.W.(1); Hayes, J.C.(1); Miley, H.S.(1); Schrom, B.T.(1). (1) Pacific Northwest National Laboratory.

Any atmospheric monitoring network of the past, today, or the future will have practical limitations in the density of sampling stations. The classical approach to measuring radioactivity in the atmosphere has been to have 12- or 24-hour integration of air samples at the highest practical station density possible to improve minimum detectable concentrations. In this work, the authors will present several considerations on optimizing the integration time and combining samples to make the best use of any network and maximize the likelihood of collecting quality samples at any given location. In particular, through the results of a number of simulation efforts this work makes the case that shorter duration sample integration (i.e. <12 hours) adds information and improves the source location capability of a network, or even just one station.

Log: 486. **ANALYTICAL REQUIREMENTS FOR DEVELOPING A NATIONAL NUCLEAR SECURITY ADMINISTRATION.** Jerry Davydov. U.S. Department of Energy / National Nuclear Security Administration.

A National Nuclear Forensics Library (NNFL) assists a State to assess whether nuclear material encountered out of regulatory control is of domestic or international origin. An NNFL collates information on domestic nuclear activities into a single or federation of databases, allowing investigators to compare information on interdicted nuclear material with domestic holdings. Much of this information already exists within a state's nuclear material registry, production history, or safeguards records, having been collected previously. By identifying the nuclear material process history, the analytical data associated with the nuclear material, and the location where the nuclear material was produced, and collating that information into an NNFL, states are left with a strong mechanism to combat illicit nuclear trafficking. The development of an NNFL begins with identifying, organizing, and populating existing nuclear material information from relevant stages throughout the nuclear fuel cycle into a single or federation of databases. In accordance with ASTM nuclear standards, as well as national and safeguards reporting requirements, miners, manufacturers, vendors, and operators routinely collect information on nuclear fuel cycle operations. In order to collect data on nuclear fuel cycle operations, these actors possess the associated technical capability required to conduct analysis of the nuclear materials they are producing or utilizing. Generally speaking, a state's nuclear fuel cycle sophistication is directly correlated with its technical capability, i.e., the more sophisticated the nuclear fuel cycle; the more advanced the technical capability. If existing information is deemed insufficient for any reason, a state could then request that the producer carry out further analyses on selected nuclear material and then utilize the new information to populate the database(s). In order to determine what technical capabilities already exist within any given state, hypothetical states are assessed based on the nuclear fuel cycle graded approach developed by the International Technical Working Group on Nuclear Forensics (ITWG). Utilizing this assessment, a state will have a better understand of the potential availability of existing information within its borders, what technical capability it possesses and may need, and ultimately the relative effort required to standup an operational NNFL.



Log: 487. **PRODUCTION OF RADIOXENON AND RADIOARGON SOURCES FOR USE IN SIMULATION OF GAS RELEASE FROM AN UNDERGROUND NUCLEAR TEST.** Haas, D.A.(1); Biegalski, S.R.(2); Bowyer, T.W.(1); Hayes, J.C.(1); Lowrey, J.D.(1); Olsen, K.B.(1). (1) Pacific Northwest National Laboratory. (2) The University of Texas at Austin.

Detection of noble gas fission and activation products resulting from underground nuclear tests is a key component of On-Site Inspections, the most confirmatory of the three verification regimes of the Comprehensive Nuclear-Test-Ban Treaty. Radioxenon and radioargon gases may reach the surface through several mechanisms, each with a characteristic time line. In order to provide empirical data on these various mechanisms, a series of tests have been carried out at the Nevada National Security Site (formerly the Nevada Test Site). These tests released tracer gases underground and require the production of operationally significant levels of radioxenon and radioargon - up to 100 Ci. The method of producing these gases through the irradiation of specially prepared targets is described in this article.

Log: 488. **A CONSIDERATION OF RADIONUCLIDE PARTICULATE RE-SUSPENSION AS A VERIFICATION TOOL IN THE CTBT ON-SITE INSPECTION VERIFICATION COMPONENT.** Lowrey, J.D.(1); Eslinger, P.W.(1); Haas, D.A.(1); Miley, H.S.(1). (1) Pacific Northwest National Laboratory.

Detection of vented radionuclides in the atmosphere comprises one major capability within the International Monitoring System (IMS) for the worldwide monitoring of nuclear explosion activity as part of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) verification regime. However, not all nuclear explosion events result in a prompt release of radionuclide gas and particulate debris to the atmosphere. In the event that an underground nuclear test does not result in measurable quantities of atmospheric radionuclides reaching IMS stations, it is still possible for a considerable amount of particulate debris to leak and be present as surface contaminant at the test region. This surface radioactivity could likely exist at levels easily detectable by one or more of the radiological methods in the possession of the CTBT On-site Inspection (OSI) Division, however, it is possible that an OSI would not be able to directly measure this surface contaminant due to the power of the Inspected States Party to block access to square restricted areas (RA) 2 km on a side. This work shows that it is possible that winds or human activity can re-suspend surface debris into the air where it could potentially be collected in high-volume aerosol samplers outside an established RA and subsequently measured in a Base of Operations (BoO) laboratory or even by particulate-detecting stations of the IMS.

Log: 489. **Generation of Gas-Phase Uranium Fluoroanions from the 1-ethyl-3-methylimidazolium Fluorohydrogenate Ionic Liquid.** Zarzana, C. A. (1); Groenewold, G. S. (1); Delmore, J. T. (1); Benson, M. T. (1); Tsuda, T. (2); Hagiwara, R. (3). (1) Idaho National Laboratory. (2) Osaka University, Graduate School of Engineering, Department of Applied Chemistry. (3) Kyoto University, Graduate School of Energy Science, Department of Fundamental Energy Science.

Fluorinating ionic liquids offer new opportunities for the synthesis of metal fluoroanions, which are of interest for fundamental structure and reactivity studies, and for use in isotope production and ratio measurements. UO<sub>2</sub> dissolved in neat 1-ethyl,3-methylimidazolium fluorohydrogenate ([EMIm][FHF]) ionic liquid to produce a blue-green solution. Analysis of this solution using electrospray-ionization mass spectrometry revealed the presence of UF<sub>6</sub><sup>-</sup> anions and clusters of UF<sub>6</sub><sup>-</sup> and EMIm<sup>+</sup>, but no other uranium-containing species. Formation of UF<sub>6</sub><sup>-</sup> required dissolution of UO<sub>2</sub>, oxidation of uranium from the +4 to the +5 state, and finally formation of the fluorouranate, all accomplished by the EMImFHF ionic liquid. The process provides a novel approach for generating stable uranium fluoroanions without using oxidizing acids like HNO<sub>3</sub>, and builds on previous research that showed that the ionic liquid stabilized HF, providing a unique synthesis environment that has the fluorinating properties of HF with less associated hazards.

Log: 490. **EFFICIENCY MODELING AND TRUE COINCIDENCE SUMMING OF THE SAGE WELL DETECTOR.** Jäderström, H (1); Mueller, W.F. (1); Atrashkevich, V (2); Adekola, A.S. (1). (1) CANBERRA Industries, Inc. (2) Consultant.

The Small Anode Germanium (SAGe) Well detector offers excellent energy resolution, a high detection efficiency for sample geometries inside the well, and a good detection efficiency for samples outside the well. The excellent energy resolution reduces the count time to reach an MDA or a given statistical precision and improves the nuclide identification because of the ability to resolve peaks close in energy. The SAGe Well detector is thus ideally suited for environmental counting applications. For the SAGe well detector, an extension to the In Situ Object Calibration Software (ISOCS) and Laboratory SOurceless Calibration Software (LabSOCS) mathematical efficiency calibration method has been developed. This allows for calculation of peak and total efficiencies for well detectors inside and outside the well. The peak and total efficiencies from ISOCS/LabSOCS have been compared to MCNP with agreements within 3% for peak efficiencies and 10% for total efficiencies for energies above 30 keV for samples inside the well. The calculation of peak and total efficiencies for the SAGe Well detector allows for calculation of true coincidence summing (TCS) using the standard Genie 2000 TCS algorithm. Due to the effect of TCS, the net peak areas of the affected full energy peaks in the spectrum suffer a loss or gain of counts. Therefore, the nuclide activity is rendered inaccurate if no correction is performed. Samples of Naturally Occurring Radioactive material (NORM), containing nuclides suffering from TCS effects have been measured with the SAGe Well detector and analyzed with an efficiency calibration from LabSOCS. Line activity consistency analysis shows a significant improvement when TCS correction is applied and the activities agree with the same sample being measured on a non-well detector.

Log: 491. **CONSIDERATION OF IMPACT OF ATMOSPHERIC INTRUSION IN SUBSURFACE SAMPLING FOR INVESTIGATION OF SUSPECTED UNDERGROUND NUCLEAR EXPLOSIONS.** Lowrey, J.D.(1); Haas, D.A.(1); Biegalski, S.R.(2). (1) Pacific Northwest National Laboratory. (2) The University of Texas at Austin.

Radioactive noble gases radon and radioargon constitute the primary smoking gun of an underground nuclear explosion. The aim of subsurface sampling of soil-gas as part of an On-Site Inspection (OSI) is to search for evidence of a suspected underground nuclear event. It has long been hypothesized that atmospheric gas can disturb soil-gas concentrations and therefore potentially add to source attribution problems in verifying treaty compliance under the Comprehensive Nuclear-Test-Ban Treaty. This work describes a study of intrusion of atmospheric air into the subsurface and its potential impact on an OSI using results of simulations from the Underground Transport of Environmental Xenon (UTEX) model. The authors draw distinction between: gas intrusion as a dilution mechanism in the subsurface, imprinting of atmospheric radionuclide source into the ground through natural ground breathing, and direct infiltration of atmosphere at the sampling point during active soil-gas sampling. Lastly, the effectiveness of installing a tarp above the sampling point in combating intrusion is evaluated.

Log: 492. **DEVELOPMENT AND OPTIMIZATION OF AN AUTOMATED URANIUM-THORIUM SEPARATION PROTOCOL FOR URANIUM AGE DATING.** Inrig, E.L.; Boulay Greene, H.; Jones, A.; Watson, I. Defence Research and Development Canada. Nuclear forensics investigations seek to reveal the origin and history of interdicted nuclear materials through the measurement and interpretation of physical, isotopic, and chemical signatures. A particularly valuable signature is the “age” of the material, defined here as the time since the last chemical purification. In the case of uranium, for all but the youngest materials this is most easily estimated using the ingrowth of Th-230 arising from the radioactive decay of U-234. Using the Bateman equations and known half-lives, the Th-230/U-234 ratio can be used to calculate the time since separation (assuming complete separation and a closed system). Particularly when the Th-230/U-234 ratio is to be determined using alpha spectrometry, complete chemical separation with high yields is of utmost importance. In this work, standard separation methods using commercially-available extraction chromatography resins and standard reagents are adapted for use with an automated radionuclide separation system. The NorthStar Engineered Technologies Automated Radionuclide Separation System (ARSIHe) – which uses a microprocessor to control a system of pumps, syringes, valves, fluid lines, and resin cartridges – is capable of rapidly executing pre-programmed separation protocols with minimal user intervention. The automated separation protocol was optimized and tested using a solution prepared from uranium and thorium mass spectrometry standards. Verification of the adequacy of the separation process and evaluation of the performance of the alpha-spectrometry-based age dating procedure as a whole is currently being carried out using a uranium certified reference material of known age.

Log: 493. **INVESTIGATION OF DYNAMIC NATURAL BACKGROUNDS FOR DISCRIMINATION IN SOIL-GAS AFTER A NUCLEAR EXPLOSION.** Lowrey, J.D.(1); Johnson, C.M.(2); Biegalski, S.R.(2); Haas, D.A.(1). (1) Pacific Northwest National Laboratory. (2) The University of Texas at Austin.

As radionuclide detection limits continue to decrease with technological advancement, it becomes increasingly important to understand the natural soil-gas background sources of radioactive noble gases radon and argon to help with source discrimination in an on-site inspection type scenario following a suspected nuclear event. In recent years, a growing emphasis has been placed on understanding the mechanisms dictating noble gas movement in the ground. In this work, the authors present simulation results of noble gas dilution and release estimates as a function of geological, meteorological, and sampling conditions using the Underground Transport of Environmental Xenon (UTEX) code. By incorporating simulated natural soil-gas backgrounds calculated in part by using MCNP's new cosmic ray source term module, post-explosion gas seepage and soil-gas sampling process in UTEX, a comprehensive examination is made of an on-site inspection scenario for study of source discrimination likelihood as well as dynamic impact of active sampling on the subsurface environment.

Log: 494. **APPLICATION FOR MODERATED D-D NEUTRON GENERATORS TO SUPPORT A RADIO-ANALYTICAL LABORATORY.** Charles K. Gary (1); Melvin A. Piestrup (1); David Diprete (2); Allan X. Chen (1); Jaakko H. Vainionpaa (3); Greg Smith (1); David W. Williams (1); Glenn Jones (4); Richard H. Pantell (5) (1) Adelphi Technology (2) Savannah River National Laboratory (3) JHV Physics (4) G&J Jones Enterprise (5) Department of Electrical Engineering, Stanford University.

Inexpensive thermal neutron sources for nuclear physics research and training are now available from Adelphi Technology Inc. Two prototype generators have been developed and have undergone preliminary testing for feasibility with neutron activation analysis (NAA). In addition, earlier models using differing moderating designs have been used for other tasks such as prompt gamma neutron activation analysis (PGNAA) and thermal neutron radiography. These sources can produce thermal neutron fluxes that are commonly achievable only by small research nuclear reactor or larger accelerator-based systems. For example, Adelphi model DD-110MB uses four Deuterium ion beams directed toward a central target and produces  $4 \times 10^7$  n/cm<sup>2</sup>-sec of thermal neutrons at the sample chamber. Advanced designs are being considered for increasing this flux to  $1 \times 10^8$  n/cm<sup>2</sup>-sec. NAA of Au and Cl samples have been demonstrated using the DD-110MB. Best geometries for source and detector placement for PGNAA and thermal neutron radiography will be discussed, along with potential issues of thermal neutron beam contamination due to fast neutrons and gamma emission. An Adelphi model DD-110MB has recently been procured by the Savannah River National Laboratory to replace an aging Cf-252 based neutron source. The neutron source is utilized to support a variety of nuclear measurement projects and radiochemical analyses. Specific applications of the generator within the radiochemical measurements and nuclear counting group will also be discussed.

Log: 495. **EXPERIMENTAL EXCITATION FUNCTIONS FOR PD-103 PRODUCED BY DEUTERON BEAMS.** Manenti, S.(1); Bonardi, M.L.(1); Gini, L.(1); Groppi F.(1). (1) Università degli Studi di Milano & INFN, L.A.S.A. Laboratory.

Brachytherapy was developed to treat prostate cancer 50 years ago. It consists in the implantation of Ti or SS seeds containing suitable radionuclides: nowadays only three radionuclides are available for use in low dose rate – LDR - prostate brachytherapy: I-125 ( $T_{1/2} = 59.4$  d, mean photon energy emitted: 21 keV), Pd-103 ( $T_{1/2} = 17$  d, mean photon energy emitted: 27 keV), and Cs-131 ( $T_{1/2} = 9.7$  d, mean photon energy emitted: 29 keV). Pd-103 is an effective alternative to I-125 for high grade, rapidly growing cancer because of its faster dose rate that also raise thanks to possible differences in external tissue complications. So it is important to study the production optimization of Pd-103 radionuclide. Nowadays Pd-103 production by accelerator is based exclusively on the irradiation of rhodium metal targets with 18 MeV protons via  $Rh-103(p, n)Pd-103$  reaction. We have studied the possibility to produce it by deuteron beams irradiation, which presents indisputable advantages. A new set of excitation functions for  $Rh-103(d,2n)Pd-103$  was measured and compared with the only other two sets present in the literature. The thin-target yields have been plotted as a function of their average energy into the targets and were fitted with the best mathematical functions. By integration of these functions the calculated thick-target yields were obtained, in order to find the optimized couple of irradiation energy and energy loss inside the thick target to maximize the production of the radionuclide of interest.

Log: 496. **UTEX SIMULATION OF NOBLE GAS MIGRATION EXPERIMENT.** Lowrey, J.D.(1); Olsen, K.B.(1); Haas, D.A.(1); Bowyer, T.B.(1); Hayes, J.C.(1). (1) Pacific Northwest National Laboratory.

In recent years, an emphasis has been placed on the need for greater understanding of the subsurface gas flow processes that can impact the potential outcome of treaty verification efforts such as on-site inspections for the Comprehensive Nuclear-Test-Ban Treaty. Numerical simulation offers one opportunity for exploring the processes that drive subsurface radionuclide migration from an underground nuclear explosion. This work aims to utilize results of a tracer gas experiment for comparison with predicted results of the Underground Transport of Environmental Xenon (UTEX) simulation code. UTEX allows for an assessment of geological, meteorological, and event-specific sensitivities, which are utilized in this study to interpret results of experimental data. The simulation results presented in this work illuminate some deficiencies in the traditional understanding of how radioactive noble gases xenon and argon move in the underground as well as the use of chemical tracer gases such as sulfur hexafluoride as surrogates in field experiments.

Log: 497. **DETECTOR READOUT ELECTRONICS WITH CUSTOMIZABLE FIRMWARE FOR HIGH-RATE LIST MODE APPLICATIONS.** Hennig, W.; Sabouarov, K.; Breus, D.; Walby, M. XIA LLC.

Digital data acquisition electronics are now commonly used to instrument radiation detectors, and can reach high rates of pulse processing with the use of on-board digital signal processors or field programmable gate arrays (FPGAs). The high rates, however, are the result of careful implementation of complex algorithms in the FPGA, not only for pulse processing as such but also for basic data handling: capturing incoming ADC data streams, transferring results into buffer memory, and sending data to the host PC. Implementation of the data handling is often outside the scope and interest of the typical end user; on the other hand, customization of the processing algorithms is often beneficial for individual applications, for example to apply specific filters for pulse height measurements or to perform detector and application specific pulse shape analysis. A recently developed electronics module, the Pixie-4 Express, uses the firmware partitioning options in modern FPGAs to provide a framework of high-rate processing firmware to which custom functions can be added. This allows users to focus on custom processing (even using higher level languages like System C), rather than be burdened with board design problems or data handling details. The Pixie-4 Express uses a 16-bit, 125 MHz ADC, a Xilinx Artix-7 FPGA, a PCI-Express host interface with several hundred MB/s sustained data transfer rate to host PC memory, and is capable of processing, buffering and storing ~500,000 events/s in list mode to a hard drive. Our presentation will describe the module's design architecture, show key firmware elements, and include results of preliminary tests in a variety of applications.

Log: 498. **VOLATILE METAL BETA-DIKETONATES FOR IMPROVED MASS SPECTROMETRIC MEASUREMENTS.** Martin, L.R.(1); Petti, M.K.(1); Johnson, A.T.(1); Finck, M.R.(1); Carney, K.P.(1). (1) Idaho National Laboratory.

The sensitivity in elemental thermal ionization mass spectrometry (TIMS) is governed by the vapor pressure of the element or compound, the ionization efficiency, and transmission characteristics of the mass spectrometer. The analysis of thorium by TIMS is made difficult due to its low vapor pressures and high ionization potential. However, the sensitivity can be improved by chemically modifying the element to increase its volatility. This has been accomplished by synthesizing various metal beta-diketonates. The volatility of lanthanide beta-diketone complexes have been well established previously through their use as precursors for vapor deposition of metal thin films. In this presentation, a solvent extraction based synthetic route for various Zr (used as an analogue for thorium) beta-diketone complexes is detailed. The resulting volatility of these complexes has been examined using thermal gravimetric analysis and IR spectroscopy. Through this analysis the viability of using this material as chemical precursors for introduction into a TIMS was determined. Ultimately the findings of this work will be applied to thorium analysis.

Log: 499. **SIMS ANALYSES OF AERODYNAMIC GLASSY FALLOUT FROM A URANIUM-FUELED NUCLEAR TEST.** Lewis, L.A.(1,2); Knight, K.B.(1); Matzel, J.E.(1); Prussin, S.G.(2); Ryerson, F.J.(1); and Hutcheon, I.D.(1). (1) Lawrence Livermore National Laboratory. (2) University of California, Berkeley.

Near-surface nuclear tests melt and vaporize large masses of soil. Much of this soil is incorporated into the nuclear fireball, where it interacts with and incorporates device debris. The resulting objects rapidly cool and solidify before reaching the Earth's surface, forming aerodynamically-shaped glassy fallout. Measuring how unfissioned fuel spatially distributes within these glasses could elucidate and constrain fallout formation processes and mechanisms. Five silicate fallout glasses produced in a uranium-fueled, near-surface nuclear test were characterized by secondary ion mass spectrometry, electron probe microanalysis, autoradiography, scanning electron microscopy, and energy-dispersive x-ray spectroscopy. The data show that individual samples display distinctive compositional heterogeneity suggestive of rapid cooling and striking heterogeneity in U isotopes. In two samples, the  $^{235}\text{U}/^{238}\text{U}$  ratio is correlated with major element composition, suggestive of the agglomeration of molten precursors. Two samples are quasi-homogeneous with respect to composition and uranium isotopic composition, suggesting extensive mixing due to slower cooling and longer retention time in the fireball.

Log: 500. **FISSION PRODUCT EXTRACTION USING DITHIOPHOSPHINIC ACID EXTRACTANTS.** D.R. PETERMAN; P.R. ZALUPSKI; R.G. McDOWELL Idaho National Laboratory.

The extraction of several fission products by the "1" / TOPO / FS-13 solvent is undesirable. Conditions necessary to minimize the extent of fission product extraction will be determined. Some of the fission products, i.e. zirconium, could likely be removed from the loaded solvent using a dilute hydrofluoric acid or ammonium fluoride scrub. Masking of fission product extraction by aqueous soluble complexants is likely more desirable than scrubbing with fluoride containing solutions. Candidate masking reagents include cysteine, methionine, and bimet. The results of the evaluation of different fission product scrubbing or stripping approaches will be discussed.



Log: 501. **EVALUATION OF RADIOACTIVE CONTAMINATION IN NUCLEAR REACTOR WASTE WATER USING GAMMA-RAY AND ALPHA SPECTROMETRY.**

Hoffman, M.K. (1); Connick, W.B. (1); Glover, S.E. (2); Spitz, H.B. (2). (1) Department of Chemistry, University of Cincinnati (2) Department of Nuclear and Radiological Engineering, University of Cincinnati.

A sample of waste water from a nuclear power plant holdup tank with input from a pressurized water reactor was evaluated for radioactive contamination. A number of activation products (e.g., Ag-110m, Co-60) and the fission product Cs-137 were detected in the water by gamma-ray spectrometry. Since the presence of Cs-137 in the water could indicate a failure in the fuel rod cladding, alpha spectrometry was then used to determine if any actinides, such as plutonium and americium, were also contaminants in the water. The particulates and colloidal matter were removed from the water using a 0.45  $\mu\text{m}$  filter, which was then dissolved with the addition of 10 dpm of Am-243 and Pu-242. The sample was prepared for alpha spectrometry using anion exchange, followed by electrodeposition. The presentation will describe

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Log: 502. **Multi-Detection Events, Probability Density Functions, and Reduced Location Area.** Eslinger, P.W.(1); Schrom, B.T. (1). (1) Pacific Northwest National Laboratory.

Several efforts have been made in the Comprehensive Nuclear-Test-Ban Treaty (CTBT) community to assess the benefits of combining detections of radionuclides to improve the location estimates available from atmospheric transport modeling (ATM) backtrack calculations. We will report a recent effort in which a Bayesian Probability Density Function (PDF) approach is used rather than a simple dilution field of regard to allow xenon and aerosol detections and non-detections to be combined mathematically. While this system is an initial test and not completed, it represents the beginning of development of a probabilistic approach to radionuclide event formation. Results of application of this method to a recent interesting radionuclide event showing a substantial reduction in the location uncertainty of that event will be presented.

Log: 503. **YET ANOTHER  $\beta/\gamma$  DETECTOR DESIGN FOR ULTRA-LOW BACKGROUND RADIOXENON DETECTION.** Warburton, W.K. (1) and Hennig, W.(1) (1) XIA LLC.

The ability to detect radioxenon isotopes in the atmosphere is critical to the Comprehensive Test Ban Treaty Organization's (CTBTO's) International Monitoring System's (IMS's) capability for providing robust verification that an underground nuclear test has occurred.  $\beta/\gamma$  coincidence detectors are currently used extensively in the IMS for this purpose and have proven to be reliable and relatively sensitive, providing detection capability at the 1 mBq/m<sup>3</sup> level measuring xenon extracted from 10 m<sup>3</sup> of air and counting for 24 hours. Recent experience, however, has shown that there would be significant advantages to being able to shorten counting times to 6 hours or less and attain sensitivities of 0.1 mBq/m<sup>3</sup>. Current  $\beta/\gamma$  detector designs are limited in this regard both by their inherent background levels and by interferences between the most common isotope Xe-133 and the metastables Xe-131m and Xe-133m. In this paper we will present a new design that uses multiple phoswich detectors surrounding the Xe sample cell and demonstrate how, by not only lowering background levels but also reducing interferences by over 90%, it offers an approach to achieving the goal of detecting 0.1 mBq/m<sup>3</sup> using only a 6 hour count and 10 m<sup>3</sup> of processed air.

Log: 504. **SEQUENTIAL EXTRACTION OF URANIUM METAL CONTAMINATION.** Murry, MM1; Spitz, HB1; Connick WB2 University of Cincinnati, 1College of Engineering and Applied Science, 598 Rhodes Hall; 2Department of Chemistry PO Box 210172 Cincinnati, OH 45221-0072.

Four 0.2 g samples collected from the compacted dirt floor of an abandoned metal rolling mill were analyzed for uranium using the NIST standard sequential extraction protocol that exposes a contaminant to a series of five increasingly aggressive solvents. The concentration of uranium in each extracted fraction can aid in predicting its eventual pathway and transport in the environment. This mill was first opened in 1910 and between 1948 and 1958 rolled uranium metal bars and rods for the U. S. Atomic Energy Commission. The building containing the mill was abandoned in the 1960s and has broken windows and a leaky roof exposing uranium and other metal contamination in the dirt floor to rain, ice, and snow. Each of the extracted fractions was traced with 10 dpm of NIST SRM 4324B 232U. Uranium was separated from each fraction using anion exchange and electroplated on a 17.50 mm diameter stainless steel planchette for alpha spectrometry analysis. Results demonstrate that approximately 75% of the uranium was extracted using ammonium acetate (solvent #2). Less than 10 % of the initial uranium content remained undissolved. A pyrosulfate fusion was used to determine the uranium concentration in the undissolved fraction. Results of this study suggests that the majority of the uranium extracted was in the hexavalent state [U(VI)], which forms highly soluble carbonate complexes at alkaline pH.

Log: 505. **DETERMINATION OF THE PU-240/PU-239 ISOTOPIC RATIO USING AN ALPHA SPECTRUM PEAK FITTING ALGORITHM.** Rim, J. H.; Tandon, L.; Porterfield, D. R.; Los Alamos National Laboratory.

This paper presents the use of a Bortels' alpha spectrum peak fitting algorithm to measure the Pu-240/Pu-239 mass ratio in plutonium samples representing different reactor burn-up. The ratio of Pu-240/Pu-239 in such materials provides important data for identifying and confirming the source/origin of the material. This ratio is typically measured using mass spectrometry and is difficult to make using alpha spectroscopy due to the close proximity of the Pu-239 and Pu-240 alpha energies. However, alpha spectroscopy is relatively simple to perform and is typically used in conjunction with mass spectrometry to measure full plutonium isotopics. This new approach makes it possible to use alpha spectroscopy to verify and confirm mass spectrometry data. It is also possible to exclusively use the peak fitting method for samples that do not require the precision of mass spectrometer for Pu-240/Pu-239 measurement. This approach will be more cost effective and save time. In this alpha method, Pu-238 peaks were first fitted and the peak shape characteristics were then used to deconvolute the Pu-239 and Pu-240 region and measure the isotopic ratio. Electrodeposition was used to prepare the uniformly coated plutonium sources for these high resolution alpha spectrometry measurements. The obtained Pu-240/Pu-239 isotopic ratios were compared to mass spectrometry data and the minimum detection limit for the alpha spectroscopy method was identified. All uncertainty calculations were performed in compliance to the Guide to the Expression of Uncertainty in Measurement (GUM). This document had been reviewed and assigned publication number: 'LA-UR-14-29518'

**Log: 506. New Approaches in Ultra-Sensitive and Ultra-High Resolution Mass Spectrometry & Application Prognosis for Radioanalytical Analysis.**

Koppelaar, David W (1); Barinaga, Charles J. (1), Hieftje, Gary M. (2), Ray, Steven (2), Marcus, R. Kenneth (3). (1) Pacific Northwest National Laboratory, EMSL. (2) Indiana University. (3) Clemson University.

New ultra-sensitive and ultra-high-resolution mass spectrometry methods are being developed and are on the horizon that have particular Interest and intrigue for radioanalytical analysis. By its atom-counting nature, mass spectrometry techniques have certain sensitivity and simplicity advantages over radiometric counting methods, and at least offer a complementary approach to difficult analytical situations. In our laboratories, we have been developing several new cutting-edge MS techniques that have analytical and/or logistical advantages for radioanalytical analyses. In the first approach we have adapted Orbitrap MS techniques for elemental and isotopic analyses. Both ICP and glow-discharge ion sources have been utilized with Orbitrap MS to achieve practical resolutions (mass resolving power) of over 300,000R. This would allow relief from many isobaric interferences in MS. A second approach is the development of an entirely new MS technique, distance-of-flight or DOFMS. This approach offers simple yet simultaneous detection of isotopes, improving both detection and isotope ratio performance over either large or small mass ranges. In addition, DOFMS provides an approach to ion collection or preparative isotope separation with interesting possibilities for combined MS-radioanalytical analysis. Finally, we are working on novel new ion sources that can be used for remote or portable analysis situations. Each of these approaches will be described with an eye towards practical radionuclear analysis applications.

Log: 507. **DETERMINATION OF FLUX AND IRRADIATION TIME IN ARCHIVED LOW-BURNUP URANIUM TARGETS.** Byerly, B. (1); Tandon, L. (1); Hayes-Sterbenz, A. (1); Martinez, P. (1); Xu, N. (1); Stanley, F. (1); Keller, R. (1); Schappert, M. (1); Thomas, M. (1); Spencer, K. (1). (1) Los Alamos National Laboratory.

Reactor conditions can be independently determined from fission product and activation product ratios. In old samples many of the short-lived fission products have decayed away necessitating the use of long lived or stable isotopes for determining reactor conditions. Xenon isotopes and daughter cesium isotopes ( $^{136}\text{Xe}/^{134}\text{Xe}$  and  $^{137}\text{Cs}/^{135}\text{Cs}$ ) can provide information on the reactor flux and ruthenium isotopes ( $^{106}\text{Ru}/^{103}\text{Ru}$ ) in conjunction with Pu isotopes can be used to infer irradiation time. Krypton isotopes ( $^{85}\text{Kr}/^{84}\text{Kr}$ ) can be used to determine cooling time. At low burnup (e.g. for weapons grade Pu production), many fission products and activation products are present at trace (sub-ppm) levels. Furthermore, many form volatile species under typical reactor operating conditions. Losses of up to 10-15% have been observed for Cs, Kr, and Xe isotopes in pressurized water reactors and boiling water reactors under normal operating conditions (1-2). Although ruthenium is not lost under typical conditions, its volatility is highly dependent on the oxidation conditions in the irradiated fuel (3). Historically these volatile fission products have been separated for isotopic determination using methods such as precipitation/coprecipitation, distillation, chromatography, anion exchange extraction, and vacuum extraction followed by sequential adsorption followed by radiochemical counting techniques. Using simple extraction chromatography methods followed by ICP-MS and TIMS analysis respectively we determined Cs, Ru (and daughter Rh/Pd), and Pu isotopes for a set of archived reactor targets that were subjected to low burn up irradiation. A gas extraction system has been developed to prepare samples for Kr and Xe isotopic on a magnetic sector gas mass spectrometer. All results will be compared to low burnup reactor models for predicting irradiation conditions. (1) Metz, V. et al. FIRST Nuclides - European Atomic Energy Community (2013). (2) Kudo, T. et al. J. Nucl. Sci. Technol. (2007). (3) Ducros, G. et al. Main findings from the VERCORS program (2005).

Log: 508. **RADON CAN BE A DETECTIVE HELPING GEOLOGISTS.** Thinova, L. (1); Johnova, K. (1); Fronka, A. (2); Rovenska, K. (2), Giammanco, S. (3). (1) CTU in Prague CR. (2) NRPI Prague CR. (3) INGV Sezione di Catania.

Extensive field measurements were conducted across varied rock types in the Czech Republic and on the slopes of Sicilian Etna, with its relatively simple rock composition, to prove usefulness of radon as a fault location indicator. Radon as an inert gas is able to travel underground even hundreds of meters under good conditions for convection transport. Thus it can be used as a tracer in obtaining information about processes in the deep parts of earth crust and in areas where tectonic faults reach surface. Revealing these hidden faults can significantly contribute to the protection of people who live in volcanic areas, or help to define areas to be avoided, before building houses in complicated tectonic and geological conditions. In both cases the health impact of high concentration of radon and associated carbon dioxide is significant. Using the high radon intensity locations, a hidden faults path could be more precisely determined, because basic geophysical methods alone are not capable to reveal the faults location with certainty. The main aim of the field work conducted was to evaluate such techniques usability in different conditions, and to select the best combination of methods for interpreting the results of soil radon and thoron concentration measurements. Comparison of the Czech Republic and the Etna results obtained, despite high population density and large altitudinal differences along the measured profiles, provided better understanding of various methods limitation and better understanding of specific radon behavior based on tectonic situation.

Log: 509. **HPLC Separation of Ho-163 from a Dysprosium Target for Neutrino Mass Measurements.** V. Mocko (1); W.A. Taylor (1); F.M. Nortier (1); J.W. Engle (1); T.E. Barnhart (2); R. J. Nickles (2); A.D. Pollington (1); G.J. Kunde (1); M.W. Rabin (1); E.R. Birnbaum (1). (1) Los Alamos National Laboratory, Los Alamos. (2) University of Wisconsin, Madison.

The rare earth isotope Ho-163, which has the lowest known Q-value (2.3-2.8 keV) of isotopes decaying via electron capture, is of interest for neutrino mass measurements with microcalorimeter detectors. Such measurements require the production of a significant amount of Ho-163 with high isotopic and chemical purity. Any other radioisotope present in a final Ho-163 based detector, irrespective of the decay path, would have a deleterious effect on Ho-163 electron capture spectra and must be minimized. In this contribution, isolation and purification of Ho-163 from a proton-irradiated dysprosium target will be discussed. A Dy metal target was irradiated with 16 MeV energy protons using a commercial dual particle cyclotron. After target dissolution, Ho-163 was separated from the bulk Dy via cation-exchange high performance liquid chromatography using 70 mM 2-hydroxyisobutyric acid at pH 4.6 as the mobile phase. Subsequent purification of the collected Ho fraction was performed to remove the 2-hydroxyisobutyrate chelating agent and to concentrate the Ho in a low ionic strength aqueous matrix. The final solution was characterized by MC-ICP-MS to determine the Ho-163/Ho-165 ratio, the residual Dy content, and total Ho-163 mass. The quality of Ho-163 product produced using two different Dy metal sources will be compared.

Log: 510. **The Ultratrace Determination of Thorium and Uranium in Fluorinated and Non-fluorinated Polymers for Use in Ultralow Background Applications.** Isaac J. Arnquist, May-Lin Thomas, Brian D. LaFerriere, Eric W. Hoppe, Mary Bliss, Jay W. Grate.

This paper discusses the methods developed for determining the radiopurity, specifically the Th and U content, of a variety of polymeric materials to be considered for use in ultralow background and rare-event physics experiments, such as dark matter and neutrino studies. This work focuses on the processes used to render the polymeric samples acid soluble for subsequent ultratrace analysis. Dry ashing methodologies using a quartz tube furnace followed by isotope dilution ICP-MS detection have been developed for two categories of polymer, fluorinated and non-fluorinated. A range of commercially-available and custom-made crucibles were screened for their effectiveness in attaining the microBq/kg (sub-pg/g) detection limits needed to meet the stringent radiopurity requirements of the ultralow background physics community. Results for a variety of polymers will be discussed.

Log: 511. **Field Alpha Spectrometry Tool (FαST) Development.** Peterson, D. S.(1); Rim, J. H.(1); Armenta, C. E.(1); Hoteling, N.(2); Plionis, A.(2). (1) Los Alamos National Laboratory. (2) Remote Sensing Laboratory.

The Field Alpha Spectrometry Tool (FαST) is a system designed to rapid field screening and analysis of samples containing alpha emitting radionuclides. The system consists of two major components; the first is a Polymer Ligand Film (PLF). The PLF enables rapid sample processing of an analyte in solid or liquid form into a planar form that is amenable to counting in an alpha spectrometer. PLFs have been shown to selectively extract actinide analytes from solution onto a substrate material. The substrate material can be directly measured with an alpha spectrometer with high resolution. The ability of PLFs to efficiently and selectively extracting certain actinides onto a substrate that facilitates measurement of high-quality spectroscopic data lends the technology as a field-based alpha measurement tool. The second part of the FαST system is an instrument designed for rapid, field-based measuring of alpha-decaying species. The instrument is designed to be operated by non-specialists and has a simple user interface. We will discuss system performance, time required to prepare a sample and collect data, and utility of the system for field-deployable applications.

Log: 512. **DEVELOPING A GUM COMPLIANT UNCERTAINTY APPROACH FOR FRAM.** Rim, J. H.; Doyle, J.; Tandon, L.; Porterfield, D. R.; Los Alamos National Laboratory.

The Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) code was developed at Los Alamos National Laboratory (LANL) to analyze gamma-ray spectra for the isotopic composition of plutonium or uranium. This software provides a means to easily process gamma-ray spectra to obtain isotopic information and is used globally for international safeguards applications. In a recent review of the FRAM code for use in nuclear forensics applications, it was determined that a more complete understanding and definition of the various sources of uncertainty that contribute to the overall uncertainty of the FRAM measurement process is required. As one of the ISO 17025 accredited, Bulk Special Nuclear Materials Analysis Program (BSAP) laboratories, LANL is required to perform an accurate and transparent uncertainty calculation on every analysis it performs. This paper describes a new approach to calculate a Guide to the Expression of Uncertainty in Measurement (GUM) compliant FRAM uncertainty to support nuclear forensics applications. This document had been reviewed and assigned publication number: 'LA-UR-14-29496'

Log: 513. **A NEW THERAGNOSTIC APPROACH IN THE MULTIFUNCTIONAL RADIONANOMEDICINE.** Groppi, F. (1); Bonardi, M.L. (1); Manenti, S. (1); Gini, L. (1); Sabbioni, E. (1,2). (1) Università degli Studi di Milano & INFN, L.A.S.A. Laboratory. (2) Aging Research Centre (CeSI), G.d'Annunzio University Foundation. Theragnostic nanomedicine is a new integrated nanotherapeutic system which can diagnose, deliver targeted therapy and monitor on-line the response to therapy. This multifunctional nanoplatform is an emerging highlight in nanomedicine that combines both diagnostic/imaging and therapeutic aspects, with a great potential for effective cancer imaging and therapy. The multifunctional/theragnostic radionanomedicine would be able to deliver the radionuclide for both imaging and therapy in a targeted manner – encapsulated nanocarriers - to cancer cells and nuclei, to improve the efficacy and safety of both cancer imaging and therapy with the help of a cancer-targeting ligand. Moreover, the development of multifunctional/theragnostic radionanopharmaceuticals is become a possible state-of-the-art in nanomedicine research. Liposomes, dendrimers, quantum dots, iron oxide, nanomicelles, perfluorocarbon and carbon nanotubes are commonly used carriers for the development of radionanopharmaceuticals. The radionuclides used in radionanomedicine for therapy are  $\alpha$ -particle emitters (Ac-225, At-211 ),  $\beta$ -particle emitters (Cu-67, I-131, Re-186, Re-188, Y-90) and Auger electron emitters (I-125, In-111, At-211 and Ga-67). Radionuclides used for imaging, are  $\gamma$  emitters (In-111, Tc-99m) and electron-particle emitters (Cu-64, F-18, I-124, Y-86, etc.). The aim of this work is to provide an overview of the current status of multifunctional radionanomedicine, and discuss issues to be addressed before exploiting the clinical benefits of radionanopharmaceuticals (radionuclide stability in the nanoradiotracers, radionuclide leakage from nanoradiotracers, possible interactions between the drug and radionuclide, nanoparticles toxicity).



Log: 514. **NOBLE GAS MIGRATION EXPERIMENT TO SUPPORT THE COMPREHENSIVE NUCLEAR-TEST-BAN TREATY VERIFICATION.** Olsen,K.B.(1); Kirkham R.R.(2); Woods,V.T.(3); Haas,D.H (4); Hayes,J.C.(5); Bowyer, .W.(6); Mendoza,D.P. (7); Lowrey,J.D. (8); Lukins,C.D.(9); Suarez,R.D.(10), Humble,P.H.(11); Ellefson,M.D(12); Ripplinger,M.D.(13); Zhong,L.(14); Mitroshkov,A.V.(15); Aalseth,C.E.(16); Prinke, A.M.(17); Mace, E.K.(18); McIntyre, J.I.(19); Stewart, T.L.(20); Mackley, R.D.(21); Milbrath,B.D.(22), Emer,D.F.(23), Biegalski,S.R.(24). 1-22 Pacific Northwest National Laboratory; 23 National Security Technology , and 2. A Noble Gas Migration Experiment (NGME) funded by the National Center for Nuclear Security and conducted at the Nevada National Security Site (NNSS) provided critical On-Site Inspection (OSI) information related to the detection of an underground nuclear explosion (UNE) event using noble gas signatures. The NGME project injected 2.49 Ci of Xe-127, 1.21 Ci of Ar-37, and 121 kg of sulfur hexafluoride, diluted in air, into a former UNE shot cavity (U-20az). These tracer gases were allowed to migrate from the cavity to near-surface and surface sampling locations and were detected in soil gas samples collected using various OSI sampling approaches. Following the injection, 168 SCUBA tanks samples were collected and analyzed for sulfur hexafluoride then for Xe-127 and Ar-37. Sixteen SCUBA tanks samples contained measurable Xe-127 and nine SCUBA tanks samples contained measurable Ar-37. Based on the detection of sulfur hexafluoride, Ar-37 and Xe-127 in the soil gas samples, we came to the following project conclusions: (1) the dilution factors calculated for sulfur hexafluoride, Ar-37, and Xe-127 demonstrate that SF<sub>6</sub> was enriched in all of the samples relative to both Ar-37 and Xe-127. (2) When Xe-127 and Ar-37 were present in a soil gas samples there were no significant differences in the Xe-127 to Ar-37 ratio in the samples relative to the ratio injected into the cavity. (3) The migratory behavior of the chemical and radiotracers did not fit typical diffusion modeling scenarios which predicted different arrival times and dilution factors for the three tracers. Results of this experiment suggest additional studies are needed to confirm the initial observations and provide a plausible explanation for the observed migration behaviors of UNE signatures as they migration from a shot cavity to surface sampling locations.

Log: 515. **THE ITALIAN CONTRIBUTION TO EXCITATION FUNCTION MEASUREMENTS FOR TC-99m PRODUCTION BY PROTON BEAMS IRRADIATION.** Manenti, S. (1); Gini, L. (1); Groppi, F. (1). (1) Università degli Studi di Milano & INFN, L.A.S.A. Laboratory.

The radionuclide Tc-99m is the most commonly used radiotracer in nuclear medicine, for diagnostic purpose, due its suitable physics characteristics:  $T_{1/2} = 6.0$  h, gamma emission at 140.5 keV suited for imaging via single photon emission computed tomography (SPECT) and causes minimal radiation dose to the patient. Since nowadays it is obtained by the Mo-99/Tc-99m generator, with the parent Mo-99 produced by nuclear reactor. The progressively closing process of the nuclear reactors around the world imposes to study alternative routes of Tc-99m production. Among them the direct production by the Mo-100(p,2n)Tc-99m reaction on highly enriched Mo-100 targets appears to be the most promising, even if this kind of production would solve only local or regional demand. In this prospective, due to some evident discrepancies among the excitation functions present in literature, some Laboratories start systematic experimental measurements of the cross sections and a careful analysis of the problems related to the direct production of Tc-99m by this route. In this contest we have contributed with new measurements and data set to the database of the cross sections, data that now appear more consistent and accurate. The thin-target yields have been plotted as a function of their average energy into the targets and fitted with the best mathematical function. Its integration gives the calculated thick-target yields, which allow finding the optimized couple of energy irradiation and energy loss inside the thick target to maximize the production. Some considerations about the long lived Tc-99g and the interfering radionuclides will be done.

Log: 516. **Biological Evaluation of Radio Nanocomposites using Au-198.** Park, J.H.; Lee, J.Y.; Yang, S.D. Radiation Instrumentation Research Division, Korea Atomic Energy Research Institute.

Nano-materials are useful in various fields of sciences, due to facile and stable fabrication method, versatile properties as well as easy size control and surface modification. Silica and gold nanocomposites have been widely used in medicinal chemistry, heterogeneous catalysis, electronics and biological sensing fields. Multifunctional hybrid nanocomposites have been synthesized by template method using a range of coupling materials. Gold radioisotopes (Au-198), which can be produced from nuclear reactor using Au-197 as a target atom, have been used for calibration source, tracer and therapeutic unsealed source. In this research, we describe the development of novel surface modified folate immobilized silica-198Au nanocomposites and its biological activities as radiopharmaceuticals. Surfactant-folate-silica-gold nanoparticles (SFSGNPs) have several attractive properties such as large surface areas, controllable particle size and the ease of large scale synthesis as well as stability. Folate-silica-gold nanoparticles (FSGNPs) are not suitable for in vivo measurements due to their low hydrophilicity, so SFSGNPs have been prepared to increase water solubility using TWEEN 60. SFSGNP prepared were characterized by XRD, SEM, TEM-EDS, Zeta-sizer, UV-Vis, Fluorescence spectroscopy and <sup>13</sup>C CP-MAS solid state NMR. Synthesized SFSGNPs were exposed in research reactor to give SFS(198Au)GNPs. The cellular uptake of SFS(198Au) were evaluated using CT-26 and SK-BR-3. The cellular uptake values were 35.17% at 15 min, 39.31% at 30 min, 41.57% at 60 min, 49.77% at 120 min for CT-26 and 32.73% at 15 min, 38.81% at 30 min, 40.57% at 60 min, 48.48% at 120 min for SK-BR-3. The biodistribution of SFS(198Au)GNPs were measured in CT-26 bearing balb/c mice at 15 and 120 min after iv injection and tumor to blood ratio was 2.18 at 120 min after injection. In this study, we described the synthesis and biological evaluation of SFS(198Au)GNPs. SFS(198Au)GNPs showed relatively high cellular uptake in a time dependent manner. The biodistribution of SFS(198Au) GNPs showed the accumulates in tumor cells at 120 min. These results suggest that SFS(198Au)GNPs has a possibilities as potential radiopharmaceuticals.

Log: 517. **Radiochemistry in support of post-detonation nuclear forensics at the National Ignition Facility.** Gharibyan, N. (1); Moody, K. (1); Despotopulos, J. (2); Grant, P. (1); Shaughnessy, D. (1). (1) Lawrence Livermore National Laboratory (2) University of Nevada, Las Vegas.

The unique neutron spectrum from thermonuclear burn of deuterium-tritium (DT) fuel, produced by the LLNL National Ignition Facility (NIF) via inertial-confinement fusion, provides a singular resource for performing basic nuclear science measurements and producing radionuclides of interest to post-det nuclear forensic investigations. The development of several experimental platforms in support of the NIF nuclear radiochemistry diagnostic has allowed fielding diverse materials at a variety of locations within the NIF chamber during high-yield DT shots. As a result, experiments have been carried out to measure the 14-MeV neutron fission yields of U-238 and Th-232 for initial benchmarking studies. Additionally, direct measurements of 14-MeV neutron cross sections were performed for target nuclides of interest (e.g., U-238 and Ir-191). The results of these primary experiments will be presented, along with an overview of current and future research plans. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Lawrence Livermore National Security, LLC

Log: 518. **HIGH-SPATIAL-RESOLUTION MEASUREMENTS OF U-ISOTOPES IN NUCLEAR FALLOUT GLASSES.** Jacobsen, B. (1); Weisz, D.G. (2); Prussin, S.G. (2); Hutcheon, I.D. (1). (1) Lawrence Livermore National Laboratory. (2) University of California, Berkeley.

Following a nuclear detonation, aerodynamic fallout glass is produced when environmental debris such as dust and loosely consolidated soil are swept into the fireball, melt, and quench. Fallout glasses also contain trace quantities of nuclear device material, e.g., unburned fuel and fission products, suggesting melted debris mixed with device material before it quenched. To understand the formation of fallout glasses, it is essential to constrain the time and temperature history of these objects, as well as understand how device material gets entrained and mixed with melted surface debris. Using the NanoSIMS 50 (Nano-Secondary-Ion-Mass-Spectrometer), a high sensitivity ion microprobe providing isotope and trace element measurements at micrometer resolution, we measured the U-isotopic composition of fallout glasses from a uranium-fueled test. Three individual small (~100 micron) glassy spheres, adhering to elongated cm-scale glassy objects, were sectioned across their mid-planes and polished flat. Uranium isotopes (<sup>235</sup>U and <sup>238</sup>U) were measured at a spatial resolution of ~500 nm with a precision of ~1.5% (2σ). The largest variations in elemental uranium concentrations are seen at the interface between the small spherical objects and the larger elongated objects. However, where the uranium concentrations at the interface show chemical diffusion profiles, no discernable diffusion profiles for U-isotopes are observed at the interface, suggesting isotopic diffusion of uranium is significantly faster than chemical diffusion of uranium. Determining the isotopic and chemical diffusion coefficients for uranium in melts of fallout composition will help constrain the time and temperature history of these objects. Such work is currently underway.

Log: 519. **EXPEDITING THE MEASUREMENT OF URINARY URANIUM BY KINETIC PHOSPHORESCENCE ANALYSIS (KPA) USING ENHANCED OXIDIZING AGENTS.** Ratner, R.T.; Nuclear Fuel Services, Inc.

Kinetic phosphorescence analysis (KPA) is a common analytical technique and provides an accurate and precise determination of urinary uranium in aqueous solutions. For bioassay samples, chemical processing of urine is required before analysis since organic constituents can interfere with KPA results. A wet-ashing digestion using hydrogen peroxide and nitric acid is conventionally used to remove interfering substances. Since the peroxide pretreatment method is an iterative process and may take up to six hours for complete oxidation of problematic samples, a more rapid and consistent digestion procedure is desired. Perchloric acid, ammonium persulfate, and potassium permanganate were each evaluated as potential oxidizing agents. Analyzed matrices were spiked with uranium standards at concentrations of 0.1 and 0.5 micrograms/L and included deionized water, synthetic urine, and composites of natural urine. Measurements were also compared to results obtained from a commercial laboratory. Sample digestion by perchloric acid proved to be costly, problematic, and more hazardous. Both ammonium persulfate and potassium permanganate methods were equally effective in the removal of organic materials, yielded excellent recoveries, and provided a more economical pretreatment procedure for uranium analysis.

Log: 520. **TRINITY FALLOUT: SPATIALLY RESOLVED ANALYSES OF MAJOR ELEMENT COMPOSITION AND PLUTONIUM ABUNDANCES.** M. Fitzgerald (1,2), K. Knight (1), J. Matzel (1), I. Hutcheon (1), K. Czerwinski (2). (1) Lawrence Livermore National Laboratory. (2) University of Nevada Las Vegas.

Recent analyses have shown Trinitite to be compositionally heterogeneous glassy material reflecting a mixture of local soil and bomb debris. This compositional diversity provides a wealth of previously overlooked information, including distinct compositional regions, flow-banding textures, and Pu correlation with both Ca and Fe. We examine Pu and major element abundances in aerodynamic fallout utilizing bulk radiometric (HPGE) and spatially resolved elemental/isotopic (EPMA and SIMS) analytical techniques for 10 aerodynamic fallout samples. Our data show tremendous variance in Pu abundance both between different fallout samples and within different compositional regions of individual fallout samples. Moreover, we find a distinct correlation between Pu and major element composition. Together with EPMA analyses of soil at the Trinity site, these observations suggest that soil composition may be a primary factor controlling the distribution of Pu in fallout glasses. Prepared by LLNL under Contract DE-AC52-07NA27344. LLNL-ABS-664916.

Log: 521. **INSIGHTS INTO THE DETERMINATION OF THE THERMODYNAMICS OF F ELEMENT SOLVENT EXTRACTION REACTIONS.** Martin, L.R.(1); Grimes, T.S.(1). Idaho National Laboratory.

Removal of the minor actinides and neptunium from used nuclear fuel raffinates continues to be an important step in fully closing the nuclear fuel cycle. There are several approaches being considered to perform this task using liquid-liquid

separations but all are governed by fundamental thermodynamic parameters. Being able to describe these processes using a thermodynamic model will be essential to eventual process deployment. However, most of the thermodynamic parameters required need to be determined experimentally, especially for the actinides. Traditionally, thermodynamic parameters for biphasic reactions have been determined indirectly using the van 't Hoff method. Although this method is sound, more recently, direct two- phase calorimetric measurements of the enthalpy of metal ion transport across the liquid-liquid boundaries has been found to be possible. In this presentation, we will discuss the differences in measuring the heat of extraction of f-elements using both the calorimetric approach and van't Hoff methodology with the merits and drawbacks being discussed.

Log: 522. **MONTE CARLO MODEL OF COMPTON SUPPRESSED DETECTORS FOR THE EXAMINATION OF SPENT FUEL** MONTE CARLO MODEL OF COMPTON SUPPRESSED DETECTORS FOR THE EXAMINATION OF SPENT FUEL. Bender, S.E. (1); Unlu, K. (1). (1) Pennsylvania State University.

The application of Compton suppression to gamma-ray measurements of spent fuel may reduce the high Compton continuum from dominant fission products to allow less intense, lower energy peaks to be detected and potentially improve the accuracy of analysis algorithms. A detector model was developed to study the application of Compton suppressed gamma-ray detection systems for the analysis of spent nuclear fuel. The Geant4 toolkit was used to simulate Compton suppressed spectral measurements of gamma radiation from spent nuclear fuel using HPGe and LaBr3 detectors. The RadSrc code, developed at Lawrence Livermore National Laboratory, was used to calculate the abundances of decay products from mixtures of heavy nuclides in addition to fission and activation products present in spent nuclear fuel to define the source term. The active detector volume and structural dimensions of the HPGe detector were confirmed experimentally to accurately simulate the detector response. The model was validated using dissolved samples of commercial fuel elements, processed with the beginning stages of the PUREX method, intended to represent the unseparated feed solution from a reprocessing facility. Compton suppressed measurements of the fuel samples were recorded inside the guard detector annulus, to simulate the siphoning of small quantities from the main process stream for long dwell measurement periods. The model shows good agreement with the measured fuel sample spectra. However, by pursuing a source definition technique that allowed for the simulation of a complete spent fuel sample, the correlation between photons emitted by the same radionuclide was lost.

Log: 523. **Thermochromatographic Analysis of Volatile Organometallic Fission Products.** Auxier II, J. D.(1), Stratz, S. A.(1), and Hall, H. L.(1). (1) University of Tennessee, Knoxville.

Solution phase separations are a fundamental process for performing radiochemical analysis. However, traditional solution based separations are generally slow, hence the need for more rapid separation techniques. This work involves the exploitation of gas-phase chemistry for the isolation of fission and activation products in the immediate aftermath of a nuclear explosion. Gas thermochromatographic separations of rare earth elements (REEs), heavy transition metals ( $Z > 72$ ), thorium, and uranium followed by subsequent detection by time-of-flight mass spectrometry (TOF-MS) will be discussed. The REEs can be readily volatilized with 1,1,1,5,5,5-hexafluoroacetylacetone (hfac), 6,6,7,7,8,8,8-heptafluoro-2,2-dimethyl-3,5-octanedione (Hfod) and 2,2,6,6-tetramethyl-3,5-heptanedione (Hdpm). in temperatures between 175 - 250 degrees C. In this work, the experimental separations as well as determination of key thermodynamic parameters that control these systems have been explored. This method offers the advantage of rapid, carrier free separations, in addition to providing elemental and isotopic composition information regarding a given sample. Furthermore, this work also supports the growing need for rapid separation and analysis of short lived isotopes currently being used in the radiopharmaceutical and nuclear medicine community and the super heavy element discovery research efforts.

Log: 524. **QUANTIFYING 240-PU IN RADIOLOGICAL MATERIAL THROUGH SPONTANEOUS FISSION GAS EMISSIONS OF RADIOXENON.** Prinke, A.M.(1); Olsen, K.B.(1); Bowyer, T.W.(1); Gesh, C.J.(1); Haas, D.A.(1); Hayes, J.C.(1); Lukins, C.D.(1); Mahoney, L.A.(1); Meacham, J.E.(2); Mendoza, D.P.(1); Reid, B.D.(1); Sevigny, G.J.(1); Sinkov, S.I.(1); Woods, V.T.(1). (1) Pacific Northwest National Laboratory. (2) CH2M HILL Hanford Group, Inc.

The objective of this project is to determine whether radioxenon air concentration measurements can be used to reliably quantify the amount of Pu-240 in radiological waste. It is often challenging to quantify the amount of Pu-240 in former plutonium processing plants and in radiological waste storage tanks. This type of radiological material inventory is traditionally done through solid sample analysis, which is often hazardous and expensive. Gas sample measurements bypass many of the hazards of traditional solid waste sampling. This project's method of quantifying Pu-240 through gas sample analysis instead of through solid samples takes advantage of one of the key signatures of spontaneous fission sources like Pu-240: radioxenon. Spontaneous fission sources produce significant quantities of the fission gases Xe-133, Xe-135, Xe-131m, and Xe-133m. A series of experiments has been undertaken to test the feasibility of using radioxenon concentrations in gas samples taken from a waste area to quantify the Pu-240 inventory of the associated waste. A Swedish Automated Noble Gas Analyzer (SAUNA) system was used to measure radioxenon concentrations of various air samples. One set of samples was taken from a controlled laboratory experiment with a well-known liquid solution containing Pu-240. A second set of samples was acquired from a Hanford Site waste tank that has undergone traditional solid sampling. A third set of samples was collected from four different decommissioned plutonium processing facilities on the Hanford Site, two of which had well-established Pu-240 inventories. The specifics of these experiments and their results will be presented.

Log: 525. **A COMPTON-VETOED GERMANIUM DETECTOR OPTIMIZED FOR LOW ENERGIES.** Stephan Friedrich, Lawrence Livermore National Laboratory.

The difficulty to detect low-energy gamma-signatures of minor isotopes due to the Compton background of high-energy radiation motivates the design of a Gamma detector with improved sensitivity at low energies. We have built such a detector by operating a thin high-purity Ge detector with a large scintillator Compton veto directly behind it. The Ge detector is thin to absorb just the low-energy radiation of interest while minimizing Compton scatter, and it employs a thin amorphous Ge electrode to minimize its dead layer. The subsequent BGO scintillator is large so that forward-scattered photons from the Ge detector interact in it at least once to provide an anti-coincidence veto for the Ge detector. For highest sensitivity, additional material in the line-of-sight is minimized, the radioactive sample is kept thin, and its radiation is collimated. The Compton-vetoed spectra show a significant reduction in the low-energy background, although not yet at the level predicted by Monte-Carlo simulations. We discuss the performance of this instrument, with an emphasis on the possibility to directly detect low-energy plutonium lines in the presence of high-energy fission products in spent nuclear fuel.



Log: 526. **CRYOGENIC GAMMA DETECTORS WITH ULTRA-HIGH ENERGY RESOLUTION ENABLE DIRECT DETECTION OF U-236 AND U-242.** Velazquez, M; Bates, C.R.; Dreyer, J.; Drury, O.B.; Friedrich, S. Lawrence Livermore National Laboratory.

Cryogenic gamma-ray detectors with operating temperatures below 0.1 K offer an order of magnitude higher energy resolution than conventional high-purity germanium (HPGe) detectors. They are sensitive thermometers that measure gamma energies from the increase in temperature upon gamma-ray absorption. We are developing gamma detectors based on superconducting transition edge sensors (TESs) and metallic magnetic calorimeters (MMCs) for non-destructive analysis (NDA) in nuclear safeguards applications. They are operated in dilution and adiabatic demagnetization refrigerators that can be cooled to temperatures between ~15 and ~100 mK fully automated and without the use of cryogenic liquids. The detectors have shown an energy resolution between ~45 and ~130 eV FWHM for gamma-ray energies below 150 keV. This resolution is sufficient to separate emissions of minor isotopes that are not visible with HPGe detectors due to strong nearby lines or due to the Compton background. Occasionally, lines are visible that we have not been able to attribute to gamma-rays from any isotope. As examples, we discuss the detection of U-236 in a skull oxide sample from uranium casting processes, and Pu-242 in mixed-isotope plutonium sample that resembles Pu in high burn-up nuclear fuel. Comparison of high-resolution NDA with mass spectrometry may be able to reduce some of the error bars for gamma-ray energies and branching ratios.

Log: 527. **ISOTOPIC SIGNATURE OF SELECTED RARE EARTH ELEMENTS FOR NUCLEAR ACTIVITIES PROFILING USING CLOUD POINT EXTRACTION AND ICP-QQQ-MS.** Lariviere, D.(1), Labrecque, C.(1) Lebed, P.J. (1) (1) Radioecology Laboratory, Laval University.

We developed a procedure for the determination of isotopic ratios of three rare earth elements (Nd, Sm, Eu). These light REE are of interest in nuclear science since they could be produced during nuclear fission, and thus can impact the natural isotopic abundance. A new cloud point extraction (CPE) method was used as a pre-concentration/separation strategy prior to measurement by inductively coupled plasma tandem mass spectrometry (ICP-QQQ-MS). To minimise polyatomic interferences in MS, the combination of decontamination achieved by CPE and the use of the reaction/collision cell in He and NH<sub>3</sub> mode was attempted and provided optimal results for the analysis of isotopic ratio in environmental samples. Isotopic ratios were initially measured in samples from San Joaquin soil (NIST-2709a), an area with little contamination of nuclear origin. Finally, samples collected from three sites with known nuclear activities (Fangataufa Lagoon in French Polynesia, Chernobyl and the Ottawa River near Chalk River Laboratory) and they all exhibited altered isotopic ratios for <sup>145</sup>/<sup>143</sup>Nd, <sup>147</sup>/<sup>149</sup>Sm, and <sup>151</sup>/<sup>153</sup>Eu. These results demonstrate the potential of the combined use of CPE and ICP-QQQ-MS for the detection of altered isotopic ratio in environmental samples applicable to nuclear anthropogenic contamination.

Log: 528. **NEW AND RAPID METHOD FOR THE DISSOLUTION OF ACTINIDES BY AUTOMATED FUSION DESIGNED FOR ICP ANALYSIS.** Lariviere, D.(1), Milliard, A.(1,2) (1) Radioecology Laboratory, Laval University. (2) Corporation Scientifique Claisse, Québec, Canada.

Acid dissolution is an effective and commonly used technique to dissolve environmental samples. This technique, however, has numerous issues, including the facts that it is time-consuming, is limited to sample with low-organic content (unless ashing is used prior to digestion), and has been shown to yield incomplete digestion for refractory elements such as actinides. A new method based on the dissolution of actinides by fusion using a M4 Fluxer (Corporation Scientifique Claisse, Québec) has been developed in our laboratory. This system uses the heat generated by propane burners to dissolve lithium borate salts which act as solvent for the analyte present in the samples. Finally, the fused sample is dropped into an acidified solution. This unique feature enables its coupling with instruments such as inductively coupled plasma (ICP). The main issue with this approach is the high lithium concentration in the samples, which is a strong interferent when using ICP instrumentation. Therefore, extraction procedures have been developed in order to separate the actinides from the sample and reagent matrix. Extraction resin chromatography, specifically TEVA and UTEVA resins (Eichrom technologies) have been used for this task. The performances of those resins in the conditions typical to lithium borate fusion have been investigated. New separation procedures have been developed in order to reduce lithium presence while quantitatively pre-concentrating the elements of interest. The overall approach proves to be quite efficient at rapidly dissolving and extracting actinides quantitatively at high yield while facilitating the measurement of these elements by ICP techniques.

Log: 529. **ANALYSIS OF PUBLIC PERCEPTION ON THE RADIOLOGICAL ACCIDENTS.** Lee, J.Y. (1); Lee, D.M. (1) Cha, M.Y (2). (1) Korea Institute of Nuclear Safety. (2) Korea Advanced Institute of Science and Technology

We examined the impact of the nuclear accident or radioactive contamination on the temporal variance in public sentiments via online social media. After the Fukushima nuclear disaster, Japan kept people safe from the physical effects of radiation but not from the psychological impact. However, the most serious damage of this accident is mental anguish of residents. Fears of radioactivity's invisible threat were entirely in SNS (Social Network Service) but the governments didn't respond to that social phenomena in their communication. SNS data during the sensitive period were interpreted at various angles by big data analytic methodologies. The results will present the direction of improvement in public communication corresponding to the fears.

Log: 530. **ANALYSIS FOR ENVIRONMENTAL TRITIUM FOLLOWING ATMOSPHERIC TRITIATED WATER EXPOSURE AROUND NUCLEAR POWER PLANT.** Lee, J.H.(1); Kim, Y.J.(1); Chae, J.S.(1); Lee, J.H.(1); Yoon, J.Y.(1). (1) Korea Institute of Nuclear Safety.

Tritium occurs in natural or industrial influence released by Nuclear Power Plants (NPPs). Heavy water reactor emitted relatively large quantities of tritium has been operated at Wolsong NPPs site. Tritium released as tritiated gas (HT) subsequently oxidizes tritiated water (HTO) in atmosphere. HTO transfers into soil or plants roots in part, and convert to the organically bound tritium (OBT). The environmental tritium finally could have an effect on the biological organisms and human body. Therefore, it is the most important to investigate tritium distribution in environment. A variety of environmental samples were analyzed for tritium in the vicinity of nuclear facilities. This study would be useful to estimate the tritium tendency in environmental and human dose effect due to radioactive material released from nuclear facilities in Korea.

Log: 531. **A comprehensive approach to uncertainty quantification in non-destructive assay.** Kirkpatrick, J.M (1); Croft, S. (2); Burr, T. (3); Venkataraman, R. (1). (1) Canberra Industries, Inc. (2) Oak Ridge National Laboratory. (3) International Atomic Energy Agency.

Thorough and accurate uncertainty quantification (UQ) is a crucial part of any physical measurement, without which the measurement result itself is of little value. This is particularly true in the field of nuclear measurements, where the over- or under-estimation of uncertainty can have high financial, environmental, and safety costs. High quality UQ is an expert task; it requires not only a comprehensive fundamental understanding of statistical analysis, but also both a detailed knowledge of the particular measurement technique for which the evaluation is being performed. Furthermore, any UQ is of limited use if it does not take into consideration the needs of the eventual end user, i.e. the consumer, of the assay results. The industry standard reference source for UQ, the ISO Guide to expression of Uncertainty in Measurement (the ISO GUM), primarily describes the application of calculational methods to a "bottom-up" approach to UQ, in which the uncertainties in measured and input parameters are estimated and then propagated in order to calculate the uncertainty on the output assay result. It emphasizes common approximation methods of statistical analysis over broader theoretical principles. Although these are necessary topics for any discussion of UQ, we believe there is an additional need for more comprehensive guidance on the subject that takes into consideration the broader context of fundamental statistical theory, application specific challenges and pitfalls, and the needs of analysis end users. We discuss the necessary elements of such a comprehensive approach; our emphasis is on the needs of practitioners in the field of non-destructive assay, but these issues have relevance to the broader nuclear measurements community.

Log: 532. **THE LEAST SQUARES FIT OF A DUAL LINEAR EQUATION USED IN DETERMINATION OF CARBON, NITROGEN, AND PHOSPHORUS IN CATTAIL USING COLD AND THERMAL PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS.** Johnson, E. (1); Zhao, L. (2). (1) School of Environment, Florida A&M University, Florida, Tallahassee, USA, (2) Harbin Blue Sky and Ocean Development of Science and Technology Ltd. Co., Heilongjiang, Harbin, P. R. China.

Previous studies, included the development of methods for the determination of carbon (C), nitrogen (N), and phosphorus (P) in cattail using cold neutron prompt gamma activation (CNPAGAA) and thermal neutron prompt gamma activation analysis (TNPAGAA) at national institute of standards technology (NIST, USA), provide a new approach that simulates a sensitivity curve for an element of interest (i.e. C, N, or P) (S), which is a function of hydrogen content (X) and disk thickness (Y) of sample as follows:  $S = aX + bY + c$  (where a, b, and c are constants). The constants a, b, and c can be obtained by the least squares fit from a group of standards with different hydrogen content and disk thickness of sample. This new approach instead of an average sensitivity used in similar researches takes the advantage of compensation of hydrogen content and disk thickness of sample. This paper describes the detailed method of the least squares fit of a dual linear equation for a sensitivity curve of an element of interest (i.e. C, N, or P) used in above studies.

Log: 533. **THE SIMULATION OF A SENSITIVITY USED IN DETERMINATION OF CARBON, NITROGEN, AND PHOSPHORUS IN CATTAIL USING COLD AND THERMAL PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS WITH THE MCNP CODE.** Zhao, L. (1); Johnson, E. (2); Landstrom, R. M. (3); Greenberg, R. R. (3); Xiao, D. Y. (4); Wang, J. F. (5); Li, G. (6). (1) Harbin Blue Sky and Ocean Development of Science and Technology Ltd. Co., Heilongjiang, Harbin, P. R. China; (2) School of Environment, Florida A&M University, Florida, Tallahassee, USA; (3) National Institute of Standards and Technology, Maryland, Gaithersburg, USA; (4) Technical Physics Institute of Heilongjiang Science Academy, Heilongjiang, Harbin, P. R. China; (5) He.

Previous studies, included the development of methods for the determination of carbon (C), nitrogen (N), and phosphorus (P) in cattail using cold and thermal neutron prompt gamma activation (CNP GAA and TNP GAA) at national institute of standards technology (NIST, USA), provide a new approach that simulates a sensitivity curve for an element of interest (S), which is a function of hydrogen content (X) and disk thickness (Y) of sample as follows:  $S = aX + bY + c$  (where a, b, and c are constants). The constants a, b, and c can be obtained by the least squares fit from a group of standards with different H content and disk thickness. Contents of C, N, and P in cattail were simultaneously determined using above sensitivity curves. Results of C, N, and P are better than the traditional method of average sensitivity and closer to traditional chemical method (i.e. elemental analyzer for C and N; the colorimetric method for P). However, it is critical for preparation of standards in practice. Therefore this paper proposes a new approach that the sensitivity can be obtained by calculating average neutron flux rate in standards using the Monte Carlo N-Particle (MCNP) code. It is compared to the sensitivity obtained in previous studies and verified by a group of cattail data that has been determined by CNP GAA and the traditional chemical method. This new approach provides a method of saving standard material and time of preparing sample and designing an analysis system, and can conduct experiments through theoretical calculation.

Log: 534. **COSMIC-RAY INDUCED RADIOACTIVE NOBLE GAS BACKGROUND CONCENTRATIONS AT HISTORIC NUCLEAR TEST SITES.** Wilson, W.H. (1); Lowrey, J.D. (2); Biegalski, S.R. (1); Haas, D.A. (2). (1) The University of Texas at Austin. (2) Pacific Northwest National Laboratory.

During an On-Site Inspection (OSI) conducted in accordance with the terms of the Comprehensive Nuclear-Test-Ban Treaty (CTBT), OSI personnel try to identify the presence of radioactive noble gases indicative of a nuclear explosion test in soil gas samples drawn at the suspected test site. Identifying the presence of radioactive noble gases in these samples could be complicated by the presence of a radioactive noble gas background produced by non-explosion sources such as cosmic-ray induced reactions in naturally occurring materials (i.e. Ca, Th, U, etc.). A suite of MATLAB scripts has been developed to support the generation of a number of high-fidelity MCNP6 models that are being used to estimate the radioactive noble gas background concentrations resulting from cosmic-ray induced reactions at several historic nuclear test sites. The MCNP6 models use cosmic-ray source terms and atmospheric and geological conditions specific to each of the test sites. Furthermore, the models include estimates of explosion debris (e.g. Pu-239) contamination concentrations applicable to each of the test sites. The principal objective of these studies was to determine if explosion debris contaminants at historic nuclear test sites might lead to substantial increases in cosmic-ray induced radioactive noble gas background concentrations at the test sites exceeding what would be expected at comparable, uncontaminated sites. Preliminary results indicate that the explosion debris contaminants can in-fact lead to elevated radioactive noble gas background concentrations that could potentially be misconstrued as resulting from a recent nuclear explosion.

Log: 535. **RADIOLOGICAL AND CHEMICAL CHARACTERIZATION OF PHOSPHO-GYPSUM: POTENTIAL HAZARDS AND SOLUTIONS.** ASHRAF E.M. KHATER, Physics Department, College of Sciences, King Saud University, Riyadh, Kingdom of Saudi Arabia, Egyptian Nuclear and Radiological Regulatory Authority, Cairo- Egypt. Phosphogypsum is a byproduct of phosphate fertilizer industry, 3-5 tones for each produced phosphoric acid. Usually, phosphogypsum contain relatively elevated concentrations of natural radionuclides such as U-238, Ra-226 and Pb-210, as well as hazardous heavy metals. Usually phosphogypsum stored in stacks. There are various possible applications of phosphogypsum in agriculture and building material industries. Phosphogypsum samples were collected from Ras Al-Khair industrial city- Saudi Arabia. Radiological (natural radionuclides specific activity-Bq/kg) and chemical characteristics of phosphogypsum were investigated using different techniques; gamma ray spectrometer based on hyper pyre germanium detector, INAA and ICP-MS. Radiological dose assessment, potential environmental impacts and possible safe applications of phosphogypsum were discussed.

Log: 536. **ENVIRONMENT IMPACTS ASSESSMENT OF IRON AND STEEL INDUSTRIES WASTES.** ASHRAF E.M. KHATER (1&2), WAFAA BAKR (2), Physics Dept., King Saud University, Riyadh- Saudi Arabia (1); Egyptian Nuclear and Radiological Regulatory Authority, Cairo- Egypt (2).

Iron and steel industry produces a huge amount of solid wastes that contain a significant concentration of hazards heavy metals and radionuclides. About 2-4 tonnes of various solid wastes (slag, sludge, dusts and scales) are generated per ton of steel production. Ore, solid wastes and soil samples were collected from four iron and steel industries that cover the different processes and production technologies of iron and steel production. Natural radionuclides such as U-238, Ra-226, Po-210, Pb-210, Ra-228 and K-40 were measured using different techniques; gamma-ray spectrometry, alpha particles spectrometry and inductively coupled plasma-mass spectrometry. The activity concentrations of some radionuclides are highly elevated, up to about 6 Bq/g. Occupation dose due dust inhalation were evaluated as well as pollution indices. The solid wastes of iron and steel industries should be completely characterization to control their potential hazards and evaluate their environmental impacts due to their disposal and/or applications.

Log: 537. **SHISHA MOASSEL SMOKING MIXTURES CHARACTERIZATION AND COMPARISON WITH OTHER TOBACCO PRODUCTS.** Ashraf E.M. Khater (1), S. Al-Hor (2), M. Amr (3), Kamal Chaouachi (4), Physics Department, College of Sciences, King Saud University, Riyadh, Kingdom of Saudi Arabia (1) , Physics Dept., Hail University, Saudi Arabia (2), Atomic Energy Authority, Cairo-Egypt (3), Fellow member of Altabacologie, Ex-DIU de Tabacologie (2006–2010), Paris XI University (4).

Cigarette smoking is a documented source of toxic trace and heavy metals. However, data about tobacco-derived products such as moassel/tabamel and jurak, used in the growingly popular shisha (narghile, hookah), have been scarce and scattered. In these conditions, the objective of this study, the first ever carried out on this scale, was to investigate the elemental contents of moassel and jurak and compare it with that of other tobacco products. Representative samples from 3 different brands were collected. Concentration of 34 elements was measured using the ICP-MS (Inductively Coupled Plasma Mass Spectrometer) as well as natural radionuclides (e.g. Ra-226, Ra-228 and K-40) using gamma ray spectrometer based on hyper pure germanium detector.. Results show that trace elements are much more abundant in cigarette tobacco than in shisha moassel. A wide range of variations was observed. For instance, the levels of As; Cd and Ni (mg kg<sup>-1</sup>) were: 1.59, 1.0 and 0.146; 1.45, 0.5 and 0.075; 3.5, 5 and 0.63; for, respectively: cigarette, moassel and jurak. Since shisha smoking is continuously targeted by antismoking groups as a “global epidemic”, a public health priority should be the design of culturally tailored products (for instance resins prepared from local plants to be mixed with the water of the pipes) based on well-established harm reduction techniques.

Log: 538. **A Chemical Separation Procedure Using Ionic Liquid Extraction For Fe-55 Quantification.** S.S. Morrison(1,2); S.B. Clark (1); S.J. Friese (3); L.A. Metz (2); B.N. Seiner (2); N.E. Uhnak (1); Z. Zhang (1).(1) Department of Chemistry, Washington State University. (2) Pacific Northwest National Laboratory. (3) Department of Chemistry, Salisbury University.

Iron (Fe) is a common component of environmental and industrial materials. When Fe is exposed to neutron sources, Fe-55 and Fe-59 activation products are formed. Quantitating these isotopes radiometrically can be difficult due to emission interferences from other activation products or from fission product contaminants. The measurement of Fe-55 activity is particularly challenging since a practical technique is low energy photon spectrometry (LEPS). Prior to LEPS analysis the bulk matrix must be removed to reduce shielding of the Fe-55 X-ray emissions. The removal of Gallium (Ga) is often a challenging task because of the similar chemical behavior of Fe and Ga. The purpose of this work was to demonstrate a separation procedure for Fe activation product analysis in a mixed sample containing both fission and activation products. The developed procedure used multiple anion exchange separations and two solvent extraction separations. The last separation in the procedure used the ionic liquid, trioctylmethylammonium salicylate to separate Fe from Ga. The procedure was evaluated using the NIST standard, 4350b (River Sediment).  $95 \pm 5\%$  of the Fe was recovered using the described separation procedure. The separation factor of Fe from Ga was greater than 6840.

Log: 539. **LIGAND-ASSISTED CAPILLARY ELECTROPHORESIS USING METHANOL-WATER MATRICIES FOR THE RAPID SEPARATION OF THE LANTHANIDE CATIONS.** Kevin Bennett and Sue Clark, Washington State University, Chemistry Department.

Ligand-assisted capillary electrophoresis is a valuable technique for rapid separation of metal ions. This technique was used to determine mobilities for trivalent lanthanides in the presence of a complexing ligand, and varying concentrations of methanol. The ligand used was alpha-hydroxyisobutyric acid (HIBA), and methanol concentrations by volume ranged from 0 to 50%. Increasing methanol concentrations were correlated with increased pKa values of HIBA, while the mobilities of the lanthanide ion-HIBA complexes decreased. Also, as an effect of the increased methanol concentration, the chromatographic resolution between metal ions generally increased. In this study, these effects have been investigated systematically to provide predictive capabilities for designing effective separations for these cations.



Log: 540. **FEEDBACK FROM OPERATIONAL EXPERIENCE OF ON-SITE DEPLOYMENT OF BIAS DEFECT ANALYSIS WITH COMPUCEA.** Berlizov, A.(1); Schachinger, A.(1); Roetsch, K.(1); Erdmann, N.(2); Schorle, H.(2); Vargas, M.(2); Zsigrai, J.(2); Kulko, A.(1); Keselica, M.(1); Caillou, F.(1); Unsal, V.(1); Walczak-Typke, A.(1). (1) International Atomic Energy Agency, Vienna, Austria. (2) EC JRC, Institute for Transuranium Elements, Karlsruhe, Germany.

The Combined Procedure for Uranium Concentration and Enrichment Assay (COMPUCEA 2nd generation) is a transportable analytical measurement system that combines the L-edge transmission and gamma-ray spectrometry for high-accuracy on-site uranium concentration and enrichment assay of uranium bearing materials (pellets, powders, scraps, solutions). After being deployed in the field, the system is operated by a team of a chemist and an NDA expert. The analytical performance of COMPUCEA meets current International Target Values guidelines for destructive assay, with a validated relative combined uncertainty of <0.4% and <0.2% for U-235 enrichment and uranium concentration, respectively. The hardware and methodology were developed at the EC-JRC-ITU and since 2007 the method has been in routine use by Euratom for physical inventory verification (PIV) at European fuel fabrication plants. In 2011 COMPUCEA 2nd generation was authorized by the IAEA and since that time it has been successfully utilized during annual PIV inspections at Ulba Metallurgical Plant in Ust-Kamenogorsk, Kazakhstan. The paper will present: (i) a summary of the establishment of a fully integrated measurement system, comprising specialized hardware and software; (ii) the results of the continued evaluation of the system's analytical performance against primary analytical methods TIMS and titration; (iii) the build-up of the ITU's and Agency's expertise in performing COMPUCEA analysis in the field. On-going effort on further extension of the system's analytical capability towards in-field analysis of UF<sub>6</sub> will be highlighted.

Log: 541. **NANOSCALE SECONDARY ION MASS SPECTROMETRY FOR NUCLEAR FORENSIC APPLICATIONS.** Kips, R.(1); Kristo, M.J.(1). (1) Lawrence Livermore National Laboratory.

Secondary ion mass spectrometry (SIMS) is an infrequently utilized, but often powerful, tool for nuclear forensic analysis. Its high spatial resolution and large dynamic range, as well as the relatively straightforward sample preparation process, make SIMS the technique of choice for measurement of isotopic and trace element abundances in inhomogeneous or particulate samples. LLNL has pioneered the use of the CAMECA NanoSIMS, an ultra-high spatial resolution, high transmission sector-field SIMS instrument, for nuclear forensic purposes. Using a finely focused beam of either oxygen or cesium primary ions, the NanoSIMS is capable of acquiring microscopic images of isotopic and elemental distributions with a nanometer-scale spatial resolution. This past year, we replaced the NanoSIMS' conventional duo-plasmatron primary ion source with an Oregon Physics Hyperion inductively-coupled plasma (ICP) ion source. The increased beam brightness of the Hyperion allowed us to reduce the spatial resolution for oxygen primary ion beams from 150-250 nm to 40 nm. We will describe the characteristics of this new ion source, as well as its application to nuclear forensics, including

results from the characterization of fuel pellets using NanoSIMS, combined with secondary electron microscopy (SEM) and energy-dispersive X-ray analysis (EDX), as part of an international round robin on the nuclear forensic analysis of fuel pellets. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory (under contract DE-AC52-07NA27344).

Log: 542. **A NANOSIMS STUDY OF THE TRANSPORT OF PU IN THE VADOSE ZONE AT THE HANFORD SITE.** Kips, R.(1); Jacobsen, B.(1); Weber, P.K.(1); Zavarin, M.(1); Felmy, A.R.(2); Kersting, A.B.(1). (1) Lawrence Livermore National Laboratory (2) Pacific Northwest National Laboratory.

The Hanford Site was established in the 1940s to produce plutonium for the US weapons program. During its operation, more than  $4.4 \times 10^{14}$  Bq of Pu-bearing liquid waste was released into the subsurface at various locations on the site. The Z-9 trench in Area 200, where the Pu finishing plant was located, received the largest volumes of acidic, high-salt transuranic liquid waste from solvent extraction processes using TBP and  $\text{CCl}_4$ . Studies have shown that Pu and other actinides have migrated as deep as 30m below the surface of the Z-9 trench. The exact transport mechanisms are still largely unknown, and more insights into the chemical behavior of Pu and the mechanisms of migration in the subsurface are needed to better assess the long-term impact of Pu transport at the Hanford Site and other locations. We characterized two soil samples from the vadose zone below the Z-9 trench in Area 200. X-ray diffraction showed that the soils were mainly composed of quartz, feldspar, clay and other silicates. Nanoscale Secondary Ion Mass Spectrometry was used to detect Pu in the soil samples. Pu was found in both samples, on the surface of several different minerals, including quartz, feldspar and Fe-oxides. The Pu was detected on many large primary minerals, as large as  $30 \mu\text{m}$ , making it unlikely that the minerals themselves were transported from the surface. We hypothesize that the Pu was not transported colloiddally, but more likely as a dissolved species in acidic waste, and precipitated out of solution at depth.

Log: 543. **RadSHARCII – a medium resolution autonomous in-situ gamma ray detection system for marine and coastal waters.** Schwantes, J.M. (1); Elster, J. (1); Addleman, R. S. (1); Colburn, H. (1); Erikson, R. (1); Fryxell, G. (2); Gervais, K. (1); Hughes, M. (2); Jokerest, T. (3); Ozanich, R. (1); Reilly, D (1); Santschi, P.E. (4); Skorpik, J. (1); Sweet, L. (1); Winterrose, M. (1); Jones, M. (1). (1) Pacific Northwest National Laboratory. (2) Canberra. (3) Texas A&M University at Galveston.

Scientists at Pacific Northwest National Laboratory (PNNL), in collaboration with researchers at Texas A&M University at Galveston, have developed a low-power, medium-resolution, autonomous in-situ gamma detection system, known as the RadSHARCII, for monitoring radionuclides in marine and coastal waters. The detection system is compatible with the Liquid Robotics Wave Glider™ platform, a wave-powered robot capable of long-term (up to one year) autonomous deployments. Two solar panels on the top of the Wave Glider provide house power to the detection system, which pumps surface water through prefilters and 75 mL chemisorbent cartridges prior to detection. Selective extraction and preconcentration of radioisotopes of interest prior to counting eliminates signal attenuation of the gamma rays traveling through water and lowers the overall background from the presence of naturally occurring radioactive isotopes (primarily from 40K and U/Th series radionuclides). Gamma ray counting is performed by a Saint Gobain 380 BrillanCe 1.5 inch LaBr3(Ce) detector coupled to a Canberra Osprey Multi-Channel Analyzer. Genie Automation and Tentative Nuclide identification algorithms are used to process spectra on-board, autonomously, and in near-real-time. On-board analysis of spectra includes energy and efficiency calibrations and an energy gain correction. Full raw spectra or a concise summary of on-board analytical results can be ex-filtrated from the system automatically in near-real-time or by command by a variety of communication methods.

Log: 544. **Analysis of Depth Profiles of B-10 and Li-6 in Si Wafers and Electrode Films of Lithium Ion Battery using KAERI-NDP System.** B.G. Park(1); G.M. Sun(2).

A Neutron Depth Profiling(NDP) technique, originally developed in 1972 by Ziegler et al, is a nondestructive near surface method that can analyze the component nuclide concentration versus depth distribution in a sample. NDP is useful for analyzing light elements with high neutron cross-section for particle-producing reactions. The NDP system has been designed and developed at HANARO, a 30 MW research reactor at Korea Atomic Energy Research Institute(KAERI). The system has been installed end of the cold neutron guide, CG1 in the Cold Neutron Activation Station(CONAS). For the performance testing and application of the KAERI-NDP system, various samples were prepared. Boron implanted and lithium deposition Si wafers and electrode films that are prepared by disassembling the a lithium ion battery used in a mobile phone were installed at the target chamber of the KAERI-NDP system and irradiated for different times with cold neutrons. The energy loss spectra of charged particles were measured using a si charged particle detector. The determined depth profiles of B-10 in Boron implanted samples were well matched with the reference values. In the case of lithium deposition samples, there was a difference in Li-6 concentration profile with SIMS method in the deep region for thick samples.

Log: 545. **ANALYSIS OF BORON IN VARIOUS CARBON MATRICES USING PROMPT GAMMA ACTIVATION ANALYSIS.** SUN, G.M.(1);KIM, J.S.(1);BAEK, HANI(1);HOANG, S.M.T.(1);CHUNG, Y.S.(1). (1) Korea Atomic Energy Research Institute.

Boron element was analyzed for various carbon matrices like graphite, graphene and CNT materials by using a Cold Neutron Prompt Gamma Activation Analysis at the HANARO Research Reactor. We prepared samples by injecting boron element into the carbon matrix along grain boundary and lattice through the thermal diffusion method. The depth profile of the boron were analyzed according to the target conditions. Peak line for prompt gammas emitted by the boron show a very peculiar shape like a trapezoid because gammas emitted by recoiling Li-7 after neutron capture of neutron by B-10 undergo a Doppler broadening. Analysis of the peak line make it possible to characterize the matrix as slowing down medium.

Log: 546. **APPLICATION OF THE ML-EM ALGORITHM IN THE SUPPRESSION OF COMPTON BACKGROUND OF GAMMA-RAYS SPECTROSCOPY SYSTEM.** Hoang, S.M.T (1); Sun, G.M (1); Kim, J.S. (1); Baek, Hani(1). (1) Korea Atomic Energy Research Institute.

In this study, the ML-EM (Maximum Likelihood Fitting by Expectation Maximization) algorithm was used to suppress the Compton background of the Gamma-rays spectroscopy system. The deconvolution of the measured gamma-rays spectrum were done based on the ML-EM method and a set of response functions of gamma-rays radiation emitted from a point source with different energy. The MCNP6 code was adopted to simulate the set of response functions in the measured energy range. The results of the ML-EM deconvolution method were compared with the experiment to estimate the performance of this method. The experiment was done with the Compton suppression system of NAA lab at HANARO (KAERI, Republic of KOREA), the photo-peak areas of deconvoluted spectra in the application of the ML-EM method were considerably raised in comparing with ones of measured spectra in the same condition. Keywords: ML-EM, HPGe, MCNP, Gamma Spectroscopy, Deconvolution.

Log: 547. **IMPROVEMENT OF THE SWITCHING SPEED OF THE IGBT BY NEUTRON IRRADIATION.** Baek, Hani (1); Sun, G.M (1); Kim, J.S (1); Ahn S.H (1); Hoang S.M.T (1). (1) Korea Atomic Energy Research Institute.

IGBT has been widely used for high power switching devices due to low on-state voltage drop and fast switching speed. But, turn-off time of the IGBT is increased by the tail-current due to stored minority carrier in n-drift region when the device is turned off. Therefore controlling lifetime technique by particle irradiation(proton, neutron etc.) has become an usefully used for production of modern power devices. In this work, IGBT devices were irradiated by fast neutrons emitted from Cf-252 up to a dose  $1E12 \text{ n/cm}^2$ . The additive energy levels within forbidden energy band were generated by the neutron irradiation. The deep energy level near or slightly higher than Fermi level are known to enhance the electrical properties of the IGBT as fast switch. The shallow levels were removed by annealing and we characterized the energy levels and measured various electrical properties like I-V, leakage current, turn-off delay time and minority carrier lifetime and so on before and after neutron irradiation. Keywords: IGBT, Neutron Irradiation, Deep Energy Level, Semiconductor.

Log: 548. **HIGH RESOLUTION MONITORING METHOD OF NEUTRON FLUX USING ACTIVATION FOILS.** KIM, J.S.(1); SUN, G.M.(2); BAEK, H.N.(3); HOANG, S.M.T.(4). (1, 2, 3, 4) KOREA Atomic Energy Research Institute.

An activation foil method has been used to monitor of neutron flux. An array of gold foils can provide 2-dimensional spatial distribution of neutron flux. In this study, a new printing method was adopted to construct neutron spatial distribution with higher resolution and give an information about the neutron energy. For that, composite metal array with several metal foils was tried. The quantitative evaluation of foil method became more difficult. The mass was measured by an XRF and volume method using confocal microscope and compared each other. The activated foils were measured by gamma-ray spectrometer and used to construct the neutron spatial distribution and energy spectrum by SAN-II code. This approach is expected to give cheaper, faster and more accurate monitoring of neutron flux and energy distribution.

Log: 549. **ALL FALLOUT IS NOT CREATED EQUAL: EXPLORATION OF MORPHOLOGICAL, CHEMICAL, AND ISOTOPIC DIFFERENCES IN TYPES OF FALLOUT FROM SINGLE EVENTS.** Knight, K.B. (1); Cassata, W. (1); Eppich, G. (1); Hutcheon, I.D. (1). (1) Lawrence Livermore National Laboratory.

Fallout from individual near-surface nuclear tests can display a striking array of morphologies. We hypothesize that a continuum of fallout formation conditions exist, and further, that fallout retrieved from a given event may represent some or all of this continuum of fallout formation mechanisms depending on key factors such as the height of burst and the immediate environment of the test. We present optical images, microscopy data, elemental data, and isotopic data from suites of fallout materials reflecting single events. Key differences within these suites of data suggest that fallout from any single event may be formed through multiple competing mechanisms, and may record different thermal, temporal, and/or chemical histories. Such differences can be exploited to probe the magnitude of thermal and chemical heterogeneities in a rapidly cooling fireball. Prepared by LLNL under Contract DE-AC52-07NA27344. LLNL-ABS-665425.

**Log: 550. MASS SPECTROMETRIC MEASUREMENT OF URANIUM ISOTOPES AND DISTRIBUTION COEFFICIENT IN FUKUSHIMA CONTAMINATED SOILS .**

Sahoo, S.K.(1) ; Mishra, S.(1,2); and Arae, H.(1). (1) NIRS Chiba Japan, (2) BARC India.

Earthquake of magnitude 9.0 followed by Tsunami on 11 March 2011 caused nuclear accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP). This resulted radioactive contamination due to deposition of mainly radiocaesium as well as many long-lived radionuclides surrounding the area. Depending upon environmental conditions, radionuclides can be mobilized in aquatic systems. Therefore, the fate and transfer of these radionuclides in the soil water system is very important for radiation protection and dose assessment. In the present study, soil and water samples were collected from contaminated areas around FDNPP. Inductively coupled plasma mass spectrometry (ICPMS) is used for total uranium concentration and thermal ionization mass spectrometry (TIMS) has been used for uranium isotopes measurement. Extraction chromatography has been used for the separation of uranium. We have observed, isotope ratio  $^{235}\text{U}/^{238}\text{U}$  is of natural origin,  $^{234}\text{U}/^{238}\text{U}$  (activity ratio) as a base line study and in a few soil samples  $^{236}\text{U}$  has been detected. For the migration behavior, its distribution coefficient ( $K_d$ ) has been determined using laboratory batch method. Chemical characterization of soil with respect to different parameters has been carried out in order to explain the radionuclide mobility in this particular area. There is a variation with uranium activity ratio where as no enrichment of  $^{235}\text{U}$  has been noticed in the studied area. However, contaminated water used for cooling the reactor vessel stored near the reactor after accident, may contain radioactive uranium. Therefore, it is of concern to understand leaching of U in the groundwater system in future.

**Log: 551. OXIDATION OF Pu(IV) IN ACIDIC SOLUTION AND STABILIZATION AT NEUTRAL pH.** Bauhn, L. (1); Hedberg, M. (2); Ekberg, C. (3). Chalmers University of Technology.

A method for stabilization of a high activity plutonium solution at neutral pH is presented. The method includes oxidation of tetravalent plutonium in acidic solution followed by carbonate complexation and neutralization. Using this method, plutonium solutions with concentrations up to 0.6 mM have been neutralized and kept stable towards precipitation for up to 2 years. The present work also includes a pre-study on the kinetics of tetravalent plutonium oxidation by ozone in acidic solution.

Log: 552. **A FACILE STRATEGY TO IMPROVE THE 14MEV NEUTRON MODERATOR IN BNCT.** Li Xianglong(1); Liu Linmao(1,2);Gu Deshan(2).(1)School of Environment Northeast Normal University.(2)School of Physical Northeast Normal University.

The main content of the paper is a three-layered moderator, which is composed of tungsten, lead, and a mixture of diamond and heavy water, was used to improve thermalization efficiency for 14MeV neutron from D-T neutron generator in boron neutron capture therapy (BNCT). In our experiments, when the thickness of each layer reached 4cm, 14cm and 21cm respectively, and the mass percentage of diamond component is 75% in the mixture layer, the thermalization efficiency reaches its summit, i.e.  $3.38 \times 10^{-4} \text{cm}^{-2}$  that prove to higher than 17% compared to previous reports. So that, the yield of the D-T neutron generator only needs to be  $2.96 \times 10^{12} \text{n/s}$  to meet the requirement of cancer treatment.

Log: 553. **IMPROVED PRECISION AND ACCURACY IN QUANTIFYING PLUTONIUM ISOTOPE RATIOS BY RIMS.** B.H. Isselhardt (1), M.R. Savina (2), A. Kucher (1), S.D. Gates (1), K.B. Knight (1), I.D. Hutcheon (1). (1) Lawrence Livermore National Laboratory. (2) Argonne National Laboratory.

Resonance Ionization Mass Spectrometry (RIMS) holds the promise of rapid, isobar-free quantification of actinide isotope ratios if laser-induced and other sources of systematic bias in the measured ratios can be overcome. We present recent progress in demonstrating this potential using two Pu test materials to demonstrate precision and accuracy in the measured Pu-240/Pu-239 ratio to about 0.3%, without any chemical purification prior to measurement. The minor isotopes Pu-238 and Pu-241 are also quantified with statistics limited precision and accuracy. In addition, we will present a preview of the new LLNL RIMS facility optimized for quantifying isotope ratios, currently under construction, and discuss the simulations performed to improve instrument design and operation. LLNL-ABS-663575.



Log: 554. **Molecular Dynamics Simulations and Experimental Studies of Tri-n-butyl Phosphate for Liquid-Liquid Extraction.** Nguyen, H.D.; Nilsson, M. University of California, Irvine, USA.

One of the most commonly used solvent extraction techniques is called PUREX (Plutonium URanium EXtraction), which employs tri-n-butyl phosphate (TBP) as an extracting agent to selectively complex Uranium and Plutonium from the constituents of the used fuel. However, our understanding of the metal extraction mechanism and phase separation involve many molecular level events are far from complete. To gain insights into the thermodynamics and kinetics of interactions of TBP molecules with other molecules in extraction systems, both molecular dynamics (MD) simulations and experiments are performed. Force field parameters for TBP have been determined by conducting a characterization study of TBP bulk liquid behavior using atomistic models with different sets of van der Waals and atomic charge parameters. Various properties of TBP in the liquid phase including the mass density, electric dipole moment, and self-diffusion coefficient are obtained and compared favorably with available experimental results. The self-association of TBPs in the presence of n-dodecane is examined by using Vapor Pressure Osmometry (VPO) technique at different temperatures, producing experiment results that are in good agreement with previous works for low concentrations of TBP/n-dodecane mixtures. However, there is a discrepancy at higher concentrations of TBP due to the assumptions of previous studies that TBP is ideal and only dimerizes in solutions. An improved model for TBP self-association is being developed by including TBP trimers and possibly higher order aggregates as observed in MD simulations. The insights gained from this study are useful for elucidating the role of TBP in phase separation.

Log: 555. **NONDESTRUCTIVE DETECTION OF 20 MICRO-BQ-ALPHA RADIOACTIVITY IN 10 GRAM SOIL OR POWDER SAMPLES BASED ON AUTORADIOGRAPHY.** Zeissler, C.J. (1).

Nondestructive analysis of soils or powders is desirable so that individual particle diameters, morphology, chemistry and discrete radioisotope content are preserved. Analysis of particles one-by-one can be too slow by microanalytical methods such as probe and microscopy techniques. This is especially true when powder or soil samples significantly exceed a milligram and the material of interest is very dilute. Autoradiography offers a nondestructive means to screen and preconcentrate relatively large quantities. For example, in 3 days, 37 grams of soil consisting of ca. 8 million soil particles was counted by autoradiography and processed to achieve a preconcentration factor of 70 consisting of 468 radioactive mineral particles (essentially zircon and monazite grains). In other words, the 468 radioactive particles in the 37g sample were located and preconcentrated into a 0.5 g sample. The result can be followed by nuclear counting such as alpha spectrometry or gamma spectrometry. A newer method using a fiducial marking system has improved the preconcentration factor further to about 110x. Redispersion and an additional autoradiograph count can be used to physically isolate the grains of interest for individual particle follow-up microanalysis by techniques such as microscopy, x-ray fluorescence and/or mass spectrometry. Soil standards and reference materials will also be discussed.

Log: 556. **NUCLEAR FORENSICS EXPERTISE DEVELOPMENT: TRANSFERRING KNOWLEDGE TO THE NEXT GENERATION.** Morrison, J.J.(1); Connelly, S.K.(2). (1) U.S. Department of Homeland Security; (2) U.S. Department of Homeland Security.

Since 2008, the National Nuclear Forensics Expertise Development Program (NNFEDP) has served as the comprehensive U.S. Government effort to grow and sustain the uniquely qualified technical expertise required to execute the nation's nuclear forensics mission. The NNFEDP has created a vibrant academic pathway over the past five years from undergraduate to post-doctorate study in nuclear and geochemical sciences directly relevant to nuclear forensics, supporting over 300 students and faculty in partnership with 11 U.S. national laboratories and 27 universities. Through its fellowship, scholarship, junior faculty, and education development initiatives, the program links next generation scientists with technical experts at the labs for practical research experiences and individual mentoring to facilitate critical knowledge transfer and to establish a seamless pipeline from academia into an attractive career in nuclear forensics. The NNFEDP provides an active and practical example of how to transfer and sustain nuclear forensics knowledge and expertise to the next generation of scientists – a major challenge facing the international nuclear security community today.

Log: 557. **ADVANCED DIFFRACTION FOR DETERMINATION OF URANIUM METAL PRODUCTION PROCESSES.** Reilly, D.(1); Arey, B.(1); Buck, E.(1); Burkes, D.(1); Schwantes, J.(1); Sweet, L.(1). (1) PNNL.

The expanding world of nuclear forensics has evolved to look at every piece of information available when answering questions about a material's past. X-ray diffraction is commonly used in nuclear material interdictions as a way to chemically identify the phase(s) present, which can help investigators determine motivation, process history, and storage history among other evidence. Although diffraction is used, it is not used to its full potential. In many cases, values such as texture, strain, and grain size orientation distribution can give more information, and possibly more specific information. In this work, electron backscatter diffraction (EBSD) with the use of a focused ion beam/scanning electron microscope (FIB/SEM) and X-ray powder diffraction data were used to evaluate signatures corresponding to uranium metal production processes.

Log: 558. **Molecular Recognition of Cesium and Francium.** Moyer, B. A., Oak Ridge National Laboratory.

As a high-yield fission product, cesium plays the dubious role as a radioactive contaminant associated with nuclear waste, nuclear accidents, and leaking storage tanks. As a result, considerable research has been devoted to the development of separation methods for cesium. Given its position at the lower left-hand side of the periodic table of the elements, cesium in its stable univalent oxidation state has the distinction of being the most charge-diffuse common metal cation. Its resulting low hydration energy in fact makes it easy to separate, and not surprisingly, a multitude of separations methods have been developed. Solvent extraction has proven useful for treating nuclear waste, and methods have employed either cation-exchange or ion-pair extraction principles. The latter tends to be weak, but it offers the advantage of stripping with water, which is ideal for downstream waste-form processing. Thus, research has focused on identifying strong cesium ion receptors. However, understanding how to accommodate the co-extracted anion has also proven to be important. Calix[4]arene-crown ethers emerged as having both the requisite binding strength and selectivity, enabling the development of a complex solvent system and extract-scrub-strip chemistry for deployment at the plant scale for the cleanup of legacy nuclear waste. The presentation will describe the fundamental chemistry of cesium ion recognition as well as aspects of its application. An extension to francium ion binding and extraction will also be discussed with potential connection with radiopharmaceuticals.

Log: 559. **SELECTIVE FLUORINATION AND SEPARATION OF METALS WITH NF<sub>3</sub> FOR MASS SPECTROMETRY ANALYSIS.** Richard A. Clark, Bruce K. McNamara, Charles J. Barinaga, James M. Peterson, David G. Abrecht, Jon M. Schwantes, and Nathan E. Ballou.

Current methods for determinations of isotopic ratios of U (and various other metals) require lengthy and complex aqueous dissolution, purification and measurement methods. Reagents used in these dissolution and purification methods may also introduce contaminants into a sample. Elemental and isotopic measurements of U are performed by mass spectrometry with thermal ionization mass spectrometry (TIMS) as the most prevalent method. Other than extensive sample preparation, the primary disadvantage of TIMS is that only one element is typically determined in a given prepared sample. We report current progress on the development of a new methodology based on the generation, separation, and measurement of volatile metal fluorides. Generation of the fluorides is accomplished through a process developed at Pacific Northwest National Laboratory (PNNL) whereby U and other elements are converted to volatile fluorides with NF<sub>3</sub>. NF<sub>3</sub> is unreactive under normal ambient conditions. At specified temperatures, NF<sub>3</sub> reacts selectively to fluorinate different species which also volatilize at specified temperatures. Utilizing these species dependent traits, elements of a sample can be sequentially, in-line separated. Metals were reacted inside a thermogravimetric analyzer (TGA), and the gas outlet was directly coupled to a quadrupole mass spectrometer (QMS) with an electron impact ionization source. Here, we present results of this project including electron ionization mass spectrum of gaseous tellurium hexafluoride and molybdenum hexafluoride.

Log: 560. **DEVELOPMENT OF SEPARATION PROCESS OF PGMS AND MOLYBDENUM FROM HIGH-LEVEL LIQUID WASTE.** Takeshita K.(1); Inaba Y.(1); Takahashi H.(1); Onoe J.(2); Narita H.(3). (1)Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology. (2) Department of Physical Science and Engineering, Nagoya University. (3) National Institute of Advanced Industrial Science and Technology.

The operation of vitrification process is disturbed by the deposition of platinum group metals (PGMs : Pd, Rh and Ru) to the bottom of glass melter and the formation of yellow phase (low viscosity glass) by molybdenum (Mo), which cause the quality reduction and the volume increase of vitrified objects. The establishment of separation technique of PGMs and Mo from high-level liquid waste (HLLW) is desired for the stable operation of vitrification process. In this study, we propose a new separation system of PGMs and Mo from HLLW, which consists of a simultaneous separation process of these metals by an inorganic adsorbent and a recovery process of them by solvent extraction technique. At first, the screening tests of inorganic adsorbent were carried out by a HNO<sub>3</sub> solution with major 7 elements (Pd, Rh, Ru, Mo, Cs, Fe and Na) and a simulated HLLW with 23 elements. We found that some ferrocyanide compounds were suitable for the simultaneous recovery of PGMs and Mo from HLLW. The recovery of PGMs and Mo from a highly-concentrated HNO<sub>3</sub> solution, which was obtained by the thermal decomposition of these ferrocyanide compounds and the acid dissolution of the decomposition product, was attainable by the combination of two extraction processes. Mo in the highly acidic solution was recovered easily by an acidic organophosphorus compound, di(2-ethylhexylphosphoric acid). PGMs were separated and recovered selectively by the application of an extraction process with thiodiglycolamide and amine type compounds.

Log: 561. **EXPERIMENTAL DERIVATION OF VOLATILE LANTHANIDE THERMODYNAMIC PROPERTIES FOR RAPID GAS-PHASE SEPARATION OPTIMIZATION.** Stratz, S.A.(1); Auxier, J.D.(1,2); Hall, H.L.(1,2). (1) Department of Nuclear Engineering, University of Tennessee, Knoxville, USA. (2) Institute for Nuclear Security, University of Tennessee, Knoxville, USA.

Modern separations of fission and activation products for forensic applications, while largely effective, are inefficient when rapid analysis of nuclear debris in a post-detonation scenario is necessary. It is therefore beneficial to explore the proficiency of gas-phase separation methods requiring significantly less time and effort during analysis. In this study, the thermodynamic variations of volatile lanthanide complexes are exploited in a Gas Chromatography Mass Spectrometer system to separate individual isotopes by elution time. Using thermodynamic laws and equations, the change in enthalpy ( $\Delta H$ ) and entropy ( $\Delta S$ ) are found for each of the volatile lanthanide complexes using experimental results. Once determined, these values can be used to optimize GCMS input parameters for maximum separation efficiency of volatile lanthanide complexes.

Log: 562. **COINCIDENCE MEASUREMENTS OF SHORT-LIVED FISSION PRODUCTS USING CYCLIC NEUTRON ACTIVATION AND DEUTERIUM TRITIUM FUSION NEUTRONS.** Pierson, B.D.; Pozzi, S.A.; Flaska, M.; University of Michigan.

Nuclear forensics is an important capability that encourages best practices in stockpile stewardship and material accountancy while deterring illegal trade of weapons-grade actinides or dangerous radioactive sources and preventing nuclear terrorism. One aspect of nuclear forensics is post-nuclear weapon detonation debris characterization. Traditionally, post-detonation debris is chemically digested and, through an assortment of measurement techniques, characterized to decipher what material was used in the bomb tamper, in the trigger, in the high explosive, what fuel was used, its impurities, what transuranics are present; all in support of the legal process of attribution. Research underway at the University of Michigan Neutron Science Laboratory by student researchers is examining singles and coincident gamma-ray data acquired from thorium-232, uranium-238, and uranium-235 starting 300 milliseconds after irradiation using a deuterium-tritium fusion neutron generator to examine the time-dependent radiation response of these actinides in a boosted nuclear weapon. From this data, independent and cumulative fission yields and time-dependent ratios of photo-peaks acquired using three high-purity germanium detectors, two 180 degrees apart and one at 90 degrees from the two, have been measured. Modeled and measured results are presented and preliminary spectra acquired from the short-lived fission products will be presented. Improvements to the statistical certainty of independent fission yields for fission events induced by fusion energy neutrons will improve the reliability of models used to predict the gamma-ray blast of a nuclear weapon. These models may be used to estimate information about bomb design in support of nuclear forensics.

Log: 563. **New approach to measurements of thoron concentration in calibration chamber.** Anita Csordás(1), Predrag Kolarž(2), Tibor Kovács(1)  
1Institute of Radiochemistry and Radioecology, University of Pannonia, Egyetem St. 10, Veszprem, H-8200, Hungary 2Institute of Physics, University of Belgrade, Belgrade, Pregrevica 118, Serbia.

In this study, 0.2 m<sup>3</sup> steel barrel is used as calibration thoron chamber with controlled ambient conditions. Constant thoron concentration is provided first by mixing thoron rich air and ambient air in pre-chamber and then introducing into the calibration chamber. Radon-thoron monitor (Sarad) is placed inside the chamber. As measuring quality insurance, ionisation properties of thoron decay products are used. High energy of  $\alpha$  particle from thoron decay (6.29 MeV) is causing massive air ionization. Energy needed for ionization of air molecules is about 32 eV, and generated ion pairs are in the size range of 0.42–1.6 nm. In clean air and controlled ambient conditions, generation of ions is constant and inflow of new ions is provided by frequent thoron decays. For ion measurements we used air ion measuring device named CDIS (CDI Ltd.). Instrument uses "Gerdien" aspiration condenser principle to measure cluster ion concentration. Method is based on the polarizing voltage applied between two aspirated coaxial electrodes where the ions of the same polarity as polarising voltage are deflected toward the collecting electrode. CDIS is placed inside the chamber so that sampled air is circulating inside and neutralizing measured ions generated from thoron, radon or other  $\alpha$  decaying products. That way only inflow of new thoron particles is measured and systematic uncertainty due to background ionization is minimized. Measurements showed consistent results and linear relationship between thoron and cluster ion concentrations. However the air ion concentration was lower than the expected according to the thoron concentration. This phenomenon may be explicable with the small volume of the chamber.

Log: 564. **4 in 1 simultaneous radon/thoron exhalation and emanation determination protocol for screening porous materials.** Z. Sas, J. Somlai, T. Kovács University of Pannonia, Institute of Radiochemistry and Radioecology, H-8200, Veszprem Egyetem str. 10 Hungary.

Under indoor conditions and underground workplaces the radon, thoron and their progenies are the main contributors of residents' natural origin radiation exposure. In order to avoid elevated radiological risk the screening of Ra-226 and Th-232 content of construction materials is not sufficient. Depending on internal structure of containing matrix exhaled radon and thoron can greatly differ, irrespectively from the natural radioisotope content. To ensure safe inbuilt the radon and thoron exhalation features has to be investigated to keep the lowest radon and thoron levels. Owing to EU BSS (2014) the indoor radon reference level is only 300 Bq m<sup>-3</sup> requires low radon exhaling building materials. However, in case of the thoron levels the authority and the scientific community have not carried the point but all side agree with that the lowest thoron levels should be kept as well. In PE-RR1 (Institute of Radiochemistry and Radioecology, University of Pannonia) an overall fast, cost effective and smart screening methodology was established to survey radiological features of porous materials focused on building materials, soils, NORM

origin industrial by-products, etc. As a result of radon and thoron exhalation measurements under free exhalation state the applied classic accumulation technique with radon monitors make the determination of specific radon and thoron exhalation rates possible. Furthermore results obtained by gamma spectrometry (Ra-226, Th-232) the radon and thoron emanation can be calculated immediately from exhalation results i.e. the applied method is a 4 in 1 technique. Several types of radon monitors with semi-conductor or pulse-ionization chambers (PIC) detectors are commercially available. One of the most common PIC detectors is the AlphaGUARD, which is manufactured by Genitron Instruments. At present more than thousand devices operate and recognized as reference device for radon measurements all over the world. Despite of numerous advantages of the AlphaGUARD the discrimination of radon and thoron has not been solved. In case of accumulation technique the disturbing effect of thoron can be determined precisely as a result of intercomparison with semiconductor detectors (e.g. RAD7) which make the thoron exhalation measurement possible simultaneously.

Log: 565. **NON-ESSENTIAL CHEMICAL ELEMENTS IN COMMERCIAL BRAZILIAN DOG FOODS.** Elias, C. (1); Fernandes, E. A. N. (1); Bacchi, M. A. (1); Bode, P.(2); Sarriés, G.A. (3); Sarriés, L. V.(4); (1) Centro de Energia Nuclear na Agricultura (CENA), Universidade de São Paulo (USP). (2) Radiation Science and Technology, Delft University of Technology (TUDelft). (3) Escola Superior de Agricultura Luiz de Queiroz (ESALQ), Universidade de São Paulo (USP). (4) Instituto de Ciências Matemáticas e de Computação (ICMC), Universidade de São Paulo (USP).

The quality of dog diets depends on adequate ingredients, free of contaminants and capable of providing optimal nutrition, for promoting long-term health. In this context, NAA was applied for assessing the presence of non-essential chemical elements in ninety five samples of dry dog food for puppies (n=32) and adults (n=63) of various brands. Both short and long irradiations were carried out, allowing the measurement of Al, As, Br, Co, Cr, Cs, La, Rb, Sb, Sc, Ti and U. These elements are not essential for dogs, while Al, Sb and U can be potentially toxic. Aluminum concentrations varied within a wide range, from < 21 mg/kg to 11900 mg/kg, with the higher values found for the same commercial brand, including super premium dog food. For Sb, concentrations ranged up to 5.43 mg/kg, being the higher values observed for six samples of dog food from one producer, but with different commercial brands. Uranium also presented higher concentrations in some commercial brands from three producers.



Log: 566. **REPRESENTATIVENESS OF SUGAR CANE CORE SAMPLING SYSTEMS.** Fernandes, E. A. N. (1); Sarriés, G.A. (2); Bacchi, M. A. (1); Bode, P.(3); Kruyff, R. (3); Dias, C.T.S. (2); Wiendl, J.A. (2); Luccas, N. (1) Centro de Energia Nuclear na Agricultura, Universidade de São Paulo. (2) Escola Superior de Agricultura Luiz de Queiroz, Universidade de São Paulo. (3) Radiation Science and Technology, Delft University of Technology.

In Brazil, the sampling of sugar cane from truckloads for quality evaluation and subsequent payment is performed in accordance to the norms and procedures recommended by CONSECANA using either horizontal or oblique core samplers. In the traditional horizontal system, the sample is composed of 3 subsamples taken with the core sampler introduced laterally in the load in 3 consecutive positions. In the oblique system, nowadays mostly used, only one sample is taken with the core sampler introduced diagonally on the surface of the load. This pilot study was conceived to assess the representativeness of both sampling systems. Two truckloads of a raw sugar cane variety from the same field were sampled at the sugar mill by doing 9 perforations with the horizontal core and 9 perforations with the oblique core. Neutron activation analysis was used to determine soil tracers (Sc, Th, Fe, Hf) in the samples to calculate the amounts of mineral impurities in the load. The average values of mineral impurities obtained by using the oblique core were greater than the horizontal core. The bootstrapping technique applied to the experimental data provided valuable information for decision making, for example showed higher variability for the horizontal core. Therefore the sampling size should be 7 and 5 for horizontal and oblique cores, respectively. Also the results confirmed the need to implement local control (blocking) for loadings.

Log: 567. **HOW RADIOACTIVE IS OUR OCEAN?.** Buesseler, K.O.(1), Durkin, C.A.(1). (1) Woods Hole Oceanographic Institution.

Our Radioactive Ocean is a crowd-funded citizen-scientist campaign that was initiated because no U.S. federal agency was (or is) currently sponsoring monitoring of ocean radioactivity in coastal waters. Yet the public is asking “is it safe” to swim, fish, use seawater in our products, and even walk the beaches along the west coast because of the concern regarding arrival of Fukushima contaminants. To answer this concern, we launched a program to engage the public in gathering samples and to provide up-to-date scientific data on the levels of cesium isotopes along the west coast of North America and Hawaii. Since January 2014 when we launched the program, individuals and groups have collected more than 50 seawater samples and raised funds to have them analyzed. The results of samples collected from Alaska to San Diego and on the North Shore of Hawaii are posted on the website <http://OurRadioactiveOcean.org>. As of the end of 2014, none of the coastal samples tested in our lab have shown any sign of cesium-134 from Fukushima (all are less than their detection limit of 0.2 Becquerel per cubic meter), though trace levels of cesium-134 are found as close as 150 km offshore. Although models suggest cesium will be at levels well below those considered of human health concern, predictions are that Fukushima derived cesium will increase in waters along the west coast of North America for the next 2-3 years, so gathering baseline data now is crucial, as will be tracking those changes in to the future. The releases from Fukushima are

ongoing, which brings with it the continued need for ocean sampling and public education on radioactivity along our coast lines on both sides of the Pacific.

**Log: 568. INFORMATION ON THE 4TH COLLABORATIVE MATERIALS EXERCISE OF THE NUCLEAR FORENSICS INTERNATIONAL TECHNICAL WORKING GROUP.**

Reilly, D.(1); Gassman, P.(1); Marsden, O.(2); Schwantes, J.(1). (1) PNNL. (2) AWE.

Founded in 1996 upon the initiative of the Group of 8 governments (G8), the Nuclear Forensics International Technical Working Group (ITWG) is an ad hoc organization of official nuclear forensics practitioners (scientists, law enforcement and regulators) that can be called upon to provide technical assistance to the global community in the event of a seizure of nuclear or radiological materials. The ITWG is supported by and is affiliated with nearly 40 countries and international partner organizations including the International Atomic Energy Agency, EURATOM, INTERPOL, EUROPOL and the UNICRI. Besides providing a network of nuclear forensics laboratories that are able to assist the global community during a nuclear smuggling event, the ITWG is also committed to the advancement of the science of nuclear forensic analysis, largely through participation in periodic table top and Collaborative Materials Exercises (CMXs). Exercise scenarios use real-world samples with realistic forensics investigation time constraints and reporting requirements. This presentation will discuss information related to the preparation and results of the 4th CMX. Due to the ongoing nature of this CMX, specific information on the exercise will be withheld until the conference.

**Log: 569. COMPARISON OF THE NUCLEAR ACCIDENTS OF CHERNOBYL AND FUKUSHIMA: THE ENVIRONMENTAL PERSPECTIVE.**

Steinhauser, G.(1); Shozugawa, K.(2); Merz, S. (3). (1) Colorado State University. (2) University of Tokyo. (3) Vienna University of Technology.

A comparison of the environmental impacts of the nuclear accidents of Chernobyl and Fukushima shows that in almost every respect, the consequences of the Chernobyl accident clearly exceeded those of the Fukushima accident. In both accidents, most of the radioactivity released was due to volatile radionuclides (noble gases, iodine, cesium, tellurium). However, the amount of refractory elements (including actinides) emitted in the course of the Chernobyl accident was approximately four orders of magnitude higher than during the Fukushima accident. For Chernobyl, a total release of 5300 PBq (excluding noble gases) has been established as the most cited source term. For Fukushima, we estimated a total source term of 520 (340–800) PBq. In the course of the Fukushima accident, the majority of the radionuclides (more than 80%) was transported offshore and deposited in the Pacific Ocean. Monitoring campaigns after both accidents reveal that the environmental impact of the Chernobyl accident was much greater than of the Fukushima accident. Both the highly contaminated areas and the evacuated areas are smaller around Fukushima and the projected health effects in Japan are significantly lower than after the Chernobyl accident. This is mainly due to the fact that food safety campaigns and evacuations worked quickly and efficiently after the Fukushima accident.

Log: 570. **IMPROVING THE REALITY OF TRAINING FOR RESPONDERS AND INSPECTORS AGAINST WMD THREATS.** Kreek, S (1); Cunningham, M (1) ; White, G. (1) ; Koncher, TR (1); Dunlop, W. (1). (1) Lawrence Livermore National Laboratory.

LLNL has been working on a concept to dramatically improve the fidelity of training for responders and inspectors across all WMD domains. Focused initially on Rad/Nuc (RN), currently higher fidelity training involves the deployment and use of RN materials or devices or their surrogates. This is expensive and also can't really be done in a manner that realistically represents the true hazards. Investing internal resources, LLNL has developed and demonstrated a prototype system that realistically reproduces true RN signals, allows the trainees to use their actual response gear and has also demonstrated a commensurate exercise management system that allows fully realistic scenarios to be played. Demonstrated for outdoor operation in which GPS tracks position initially, we also have a concept for furthering the capability to allow for high fidelity use indoors. And, with the integration of our demonstrated ultrawide band communications capability, would enable training in virtually any exercise venue (ships, high rise buildings, etc). We have been discussing the vision with a number of actual responders and they are very receptive. We have also been talking under NDA with a number of instrument manufacturers in the hopes that they will design their next generation instruments to be compatible with our vision. This presentation will describe the capability demonstrated to date. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Lawrence Livermore National Security, LLC, LLNL-PRES-657133.

Log: 571 **NEW INFORMATION REGARDING THE INTERACTION OF PUEBLOAN VILLAGES OF NEW MEXICO.** James, W.D. (1), Eckert, Suzanne L. (1), Habicht-Mauche, Judith (2). (1) Texas A&M University, (2) University of California Santa Cruz.

Researchers have long sought to understand the interaction of ancient peoples from the West-Central New Mexico region. In our previous work in which we studied the chemical composition of materials collected from the Tijeras Pueblo of the Central Rio Grande region, we found evidence of distinctive glazed pottery that did not originate from the compared Zuni western site nor with local wares thought to have been made in the Albuquerque area. It was postulated that these materials may have been imported from the western Acoma site but sampling has not traditionally been allowed from this site. Recently our group was allowed to collect 125 pottery sherds from the Acoma site with approval to perform neutron activation with the hope of clearing up this issue. A description of the neutron activation analysis procedures and the results, including multidimensional grouping of sherds represented by these new data and a comparison to our previous work and literature data will be provided.

Log 572. **USING NEUTRON ACTIVATION ANALYSIS TO TRACK BRAZILIAN BEEF.** DE NADAI FERNANDES, E.A.(1), BACCHI, M.A.(1), SARRIÉS, G.A.(2), GONZAGA, C.L.(1), SARRIÉS, S.R.V.(1), FOGAÇA, A.A.(1), REYES, A.E.L.(2), BORTOLETO, G.G.(3)  
1. Nuclear Energy Center for Agriculture, University of São Paulo, 2.

Brazil occupies the first position in the world for beef exports and second in beef production and consumption. In more recent years, food safety issues have become much more prominent in the beef supply chain since consumer confidence has been undermined by epidemics such as bovine spongiform encephalopathy as well as foot and mouth disease. Legislation has increased restrictions on beef trade worldwide. In the European Union, beef trade requires traceability throughout the supply chain, allowing the customers to be aware of the origin of the meat available in the market. The traceability is usually based on the labeling and information technology, rather than the intrinsic characteristics of the meat. Analytical methods determining essential and non-essential chemical elements are potential tools to establish the geographical origin of the meat, since certain regions may provide specific fingerprints, especially trace elements. Thereby, instrumental neutron activation analysis was used to chemically characterize beef samples from the main producing areas of Brazil, in the states of Goiás, Mato Grosso, Minas Gerais, Para e Rio Grande do Sul. For this evaluation, a comprehensive sampling of the most prized cut of meat in the country, which is picanha or rump cap, with the specified origin was performed in the local market of Picacicaba. Among the elements determined, Br, Cs, and Rb showed the greater variability among the samples and could separate them according to their origins with the 95% confidence. Additional chemical elements should be investigated to improve robustness and applicability of the proposed methodology for tracking beef origin.

Log 573: **226Ra ANALYSIS FROM NATURALLY OCCURRING RADIOACTIVE MATERIALS BY LSC.** Jung, Y.(1)(2); Ji, Y.Y.(1); Lim, J.M.(1); Ko, Y.G.(1); Chung, K.H.(1); Suh, K.S.(1)(2); Kim, H.(1). (1) Korea Atomic Energy research Institute. (2) Korea University of Science and Technology.

226Ra analysis is of particular interest for radiation protection, environmental and geological purposes. It is produced and accumulated in the environment as a result of industrial activities such as phosphate industry, oil and gas extraction industries. In worldwide, the radioactivity of 226Ra in raw materials and by-products has been controlled by national regulators. Several analytical techniques have been applied to determine 226Ra in liquid and solid materials. In this study, we described the analytical method to determine 226Ra in raw materials/by-products, which was established with 222Rn emanation technique by LSC. 226Ra is extracted from solid powder samples by fusion with LiBO<sub>2</sub> (K2 PRIME, Katanax). 226Ra is co-precipitated as Ba(Ra)SO<sub>4</sub> and it converts to Ba(Ra)CO<sub>3</sub>. It is dissolved in 3% HNO<sub>3</sub> and prepared for LSC analysis. Maxilight (Hidex) was used as a cocktail, which is used for 222Rn extraction. 133Ba was used as tracer and simultaneously analyzed with 222Rn and its decay progeny by LSC. The developed method was validated with SRM 4357, 388, 600 and 1633c. The recovery of it ranges 70 % to 90 %.

**Log 574: ATMOSPHERIC DISPERSION AND IMPACT ASSESSMENT MODELLING SUPPORT IN AN EMERGENCY REGARDING CIVILIAN PROTECTION ISSUES.**

Benamrane, Y. (1); Armand, P. (1); Wybo, J-L.(2). (1) French Alternative Energies and Atomic Energy Commission. (2) Mines ParisTech

Catastrophic events such as Bhopal (1984), Chernobyl (1986) or Fukushima (2011), underline the potential large scale of damages due to radiological or chemical atmospheric releases. Nowadays, health consequences of such releases are assessed using atmospheric dispersion modeling and decision-support systems. These systems provide the first responders and decision makers with the space and time distribution of the toxic species, the radiological or chemical exposure, thus the areas where population protection measures are to be taken. We argue that modeling and decision-support systems play an increasing role in emergency management reinforced by the nuclear field experience, especially Chernobyl and Fukushima accidents. Twenty-five years apart, these systems have reached a technical maturity allowing them to be integrated in nuclear crisis centers and used in situ as operational diagnostic and / or prognostic tools. Although modeling and decision-support tools have reached a high scientific quality, their use in emergency situation remains a great challenge in providing efficient support to stakeholders in the first hours and days of the crisis, characterized by its uncertain and changing environment, high stakes and the time pressure. In this paper, we study the use of modeling tools dedicated to atmospheric dispersion and impact assessment by organizations in charge of emergency management. The analysis is based on stakeholders' interviews and observations of the interactions between scientific advisers and decision makers during training exercises implying toxic releases. Strengths and limitations in the use of modeling systems are commented on as the way to improve their effective use in an emergency.

**Log 575: GAMMA SPECTROSCOPY ANALYSIS OF ARCHIVED MARSHALL ISLAND SOIL SAMPLES.**

Herman, S (1); Hoffman, K. (1); Trauth, A. (1); Lavelle, K. (1); LaMont, S.P. (1, 2); Hamilton, T (3); Glover, S.E. (1); Connick, W. (1); Spitz, H. (1). (1) University of Cincinnati, (2) Los Alamos National Laboratory, (3) Lawrence Livermore National Laboratory.

Radiochemical analysis of samples collected from North Pacific atmospheric nuclear testing sites is complex because radioactive material may be bound to the sample media in different or unusual chemical forms. Four samples of archival Marshall Islands soil were subjected to non-destructive, broad energy (17 keV – 2.61 MeV) gamma-ray spectrometry analysis using a series of different HpGe detectors. These archival samples were collected in 1967 from different locations on Bikini Atoll affected by different weapons tests. Initial results from all samples show a range of radionuclide content that includes plutonium, <sup>241</sup>Am, and <sup>137</sup>Cs. Some samples also contained <sup>60</sup>Co and <sup>232</sup>U. Unlike samples collected recently from the Marshall

Islands, these archived samples exhibit characteristics that have been unaffected by approximately 50 years of additional weathering or other modes of environmental transport that could significantly alter critical signatures. These non-destructive gamma-ray spectrometry analyses have been used for training and to develop new experiments to evaluate the radionuclide signatures as a necessary first step prior to investigating the environmental chemistry of radionuclides in historic and recently collected samples.

**Log 576: PRECONCENTRATION OF ACTINIDES FROM CALCIUM RICH ENVIRONMENTAL SAMPLES COLLECTED FROM FORMER PACIFIC ISLAND NUCLEAR TEST SITES.** Hoffman, K.(1); Trauth, A.(1); Herman, S.(1); Lavelle, K.(1); LaMont, S.(2,3); Hamilton, T.(3); Glover, S.(1); Connick, W.(1); Spitz, H.(1). (1) University of Cincinnati, (2) Los Alamos National Laboratory, (3) Lawrence Livermore National Laboratory.

Forensics analysis to determine chemical and radiological signatures in samples of post-detonation debris may be complicated by the high concentration of calcium, iron, manganese, and other naturally occurring minerals that must be separated from the sample matrix before actinide analysis can be performed. Eichrom Technologies' DIPEX® Actinide Resin (DIPEX) has been used to eliminate high concentrations of iron in samples of rusty metal and to separate calcium in soft tissue and bone for analysis of plutonium and uranium. This resin is capable of strongly chelating tri- and tetravalent actinides on its diphosphonic acid groups while allowing elution of unwanted matrix constituents. DIPEX pre-concentration is therefore well suited for matrix elimination prior to chemical separations. This research presents a DIPEX based pre-concentration method for the determination of uranium, thorium, plutonium, americium and curium in samples with high concentrations of calcium and other interfering minerals. The method was evaluated using a range of samples of different mineral content, including Marshall Island Soil, to determine recovery and identify any significant limiting conditions. The radioactivity and mineral content of Marshall Island soil has been well characterized in the archival literature. Its high calcium content makes Marshall Island soil a good natural matrix simulant for analysis of actinides in concrete and construction debris.

**Log 577: COMPTON BACKGROUND SUPPRESSION WITH A MULTI-ELEMENT SCINTILLATION DETECTOR USING HIGH SPEED DATA ACQUISITION AND DIGITAL SIGNAL POST PROCESSING.** Popescu, G.; Glover, S.E.; Spitz, H. University of Cincinnati.

A multi-element scintillation detector, consisting of eight coupled NaI(Tl)/BGO scintillators surrounding a single coupled NaI(Tl)/CsI(Tl) dual scintillator, was previously developed for locating and measuring concealed special nuclear materials that emit primarily low energy photons. The BGO scintillators provide active collimation for the center detector. Thin (3mm) NaI(Tl) scintillators on the

faces of the BGO and CsI(Tl) scintillators detect low energy photons from uranium, plutonium, and  $^{241}\text{Am}$  while the BGO scintillators provide active collimation for the single central CsI(Tl) detector. Recently, high speed multi-channel signal digitizers and LabView virtual instrument (VI) subroutines have been used to post process and analyze signals from each of the dual scintillators to suppress Compton interactions in the central detector as a means to significantly improve the signal-to-noise ratio for gamma spectroscopy. Compton suppression is attained by rejecting events in the central detector whenever a coincident event is detected in any one of the surrounding eight dual-scintillator detectors. Use of high-speed multi-channel signal digitizers and LabView VIs for pulse post processing eliminates the classic use of analog signal processing and anticoincidence gating to reject Compton events for background reduction. The suppressed and unsuppressed pulse energy spectrum can easily be displayed for analysis. This multi-element scintillation detector operated with Compton suppression now has extended applications in aerial surveillance and environmental monitoring soil for special nuclear materials and radionuclides that emit higher energy photons.

Log 578: **POTASSIUM IN AGROFORESTRY SOILS MEASURED BY GAMMA-RAY SPECTROMETRY AND NEUTRON ACTIVATION ANALYSIS.** BIGUZZI, F.A. (1); DE NADAI FERNANDES, E.A.(1); BACCHI, M.A.(1); SARRIÉS, G.A.(2); REYES, A.E.L.(2); GONZAGA, C.L.(1). 1. Nuclear Energy Center for Agriculture, University of São Paulo; 2. College of Agriculture Luiz de Queiroz, University of São Paulo

Potassium has two natural stable isotopes ( $^{39}\text{K}$  and  $^{41}\text{K}$ ) and one natural radioisotope ( $^{40}\text{K}$ ). The measurement of  $^{40}\text{K}$  by gamma-ray spectrometry has been successfully used for calculating total K content in different types of matrices, assuming that there is no significant variation in the isotopic abundances. Here, an analytical procedure for assessing total K in soils samples by measuring  $^{40}\text{K}$  was evaluated, comparing the results to those obtained by NAA. Twelve samples of agroforestry soils collected in the Atlantic Forest, São Paulo state, Brazil, were air dried and weighed (2 kg) into Marinelli containers. The  $^{40}\text{K}$  activity was measured for 1 hour using a HPGe detector with 50% relative efficiency at 1332 keV  $^{60}\text{Co}$ . Results of total K from both methods showed a strong correlation ( $r = 0.9729$ ) with maximum relative deviation of 11%. The differences between the results were all covered by the measurement uncertainties. Zeta scores varied from 1.45 and 1.32, well within the acceptable range. The measurement uncertainties were similar for both methods, while the detection limits were 12 times lower for the  $^{40}\text{K}$  method, reaching as low as 0.02%. Based on such results,  $^{40}\text{K}$  can be used for determining total K in soil samples, despite the huge discrepancy on the abundance of  $^{40}\text{K}$  recently reported in Japanese and Slovenian forest soils. The  $^{40}\text{K}$  method can be advantageous compared to other analytical options since it is purely instrumental, with no chemical procedures, and uses a large sample providing better representativeness.

Log 579: **MULTICOLLECTOR ANALYSIS OF PICOGRAM LEVEL URANIUM ISOTOPES USING FULL SIZE MULTIPLIERS.** Black, C.K.(1); Roberts, D.(1); Burrows, A.(1); Freedman, P. (1) Nu Instruments Limited.

Simultaneous measurement of uranium isotopes is a technique that has been used for many years in mass spectrometry to minimize the loss of sample and increase measurement precision. Usually the measurement is performed using large samples with Faraday buckets. Small sample work has also previously been performed using miniature ion counters. The Nu TIMS instrument with the Nu patented zoom lens system can be equipped with five full size ion counters as well as 16 Faraday detectors. This enables simultaneous measurement of either large samples on the Faraday detectors or small size samples on the ion counters with no movement of detectors. The zoom lens system ensures excellent alignment of peaks with little to no distortion. It also provides the means to measure with full size tried and tested ion counters for the ultimate in accuracy and precision. The data presented in this poster will be a demonstration of the flexibility of the Nu TIMS instrument for low level uranium analysis of nuclear measurements.

Log 580: **LA-ICP/MS ANALYSIS OF PLASTICS AS A METHOD TO SUPPORT POLYMER ASSAY IN THE ASSESSMENT OF MATERIALS FOR LOW BACKGROUND DETECTORS.** Grate J.W. (1); Bliss M. (1); Farmer III O.T. (1); Thomas M.L. (1); and Liezers M. (1). (1) PNNL

Ultra low-background radiation measurements are essential to several large scale physics investigations, such as those involving neutrinoless double beta decay, dark matter detection (such as SuperCDMS), and solar neutrino detection. There remain needs for electrically and thermally insulating dielectric materials with extremely low background radioactivity to be used in the construction of the detectors, a need best met with plastics. There is a need to move from materials with milliBq/kg levels toward those with low microBq/kg levels. However, the assay of polymer materials for extremely low levels of radioactive elements, U and Th in particular, presents new challenges. It is only recently that any certified reference materials for toxic metals, such as Pb or Cd, in plastics have become available. However, there are no certified reference materials for U or Th in thermoplastics. In this talk, we assess the use of laser ablation for sampling and analysis of plastic samples with emphasis on U determination. Using a Certified Reference Material for Pb in polyethylene (PE), we examine laser ablation and ICP/MS parameters that determine whether the total atom efficiency for U and Pb are similar, and explore methods to use the Pb content in a plastic as part of the process of estimating or determining the U content by LA-ICP/MS.



Log 581: **FISSIONABLE PARTICLE IDENTIFICATION AND ANALYSIS FOR TREATY VERIFICATION AND NUCLEAR FORENSICS.** Morey, M. (1), Manard, M. (1); O'Neil, M. (1); Russo, R. (2); Quarles, D. (2); Gonzales, J. (2); Oropeza, D. (2); East, L. (2); Chiriros, J. (2); Yoo, J. (2); Havrilla, G. (3); McIntosh, K. (3). (1) National Securities Technology/ Department of Energy; (2) Applied Spectra/Lawrence Berkely National Laboratory; (3) Los Alamos National Laboratory

Since the release of the genie from the bottle decades ago followed by the end of the Cold War, the threat of nuclear nonproliferation and risk of nuclear terrorism has heightened significantly. An appropriate response to such actions requires quick analysis and screening of trace evidence in the field to determine attribution. This forensic can include the bomb type (Pu or U) and isotopic ratios which could give clues to the fissile material's origin or yield. Advances in laboratory and potentially fieldable analytical tools have made rapid sample attribution on location closer to reality. In this study, we report the methodology development and results of applying orthogonal and complementary techniques to the study of Trinitite and related surrogates. We have employed more established analytical techniques such as Raman, XRF, Laser Ablation (ICPMS, Ion Mobility MS), LIBS, with the recently reported Laser Ablation Molecular Isotopic Spectrometry (LAMIS) and a picoliter deposition system (TIPS). TIPS samples enabled us to calibrate the LA-ICP-MS over 6 orders of magnitude of sample concentration.